Nova Scotia Research Foundation Corporation
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TECHNOLOGY IN ACTION

# CATALYTIC TWO-STAGE LIQUEFACTION

# OF NOVA SCOTIA COALS

## FINAL REPORT

by

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## SUMMARY

The first part of a research program on catalytic two-stage liquefaction of Nova Scotia bituminous coals has been completed. Actual work was concentrated on testing three Sydney area coals with a nickel-molybdenum catalyst. Experiments were carried out in a once-through processing mode using a continuous-flow bench unit. In addition, the planned extension of a two-stage thermal/catalytic liquefaction run with liquid recycle has been completed.

During the previous phase of work, the last run was a thermal/catalytic recycle liquefaction test of six days duration using Prince Hub seam coal. The extension of that test was halted after about five days due to unfavourable experimental results. As the run progressed the donor capacity of the recycle solvent decreased, liquid product yield fell off and product quality declined. Coal conversion decreased from 93 to 85% over the eleven-day operating period. The net $C_4^+$ liquid yield during the last twelve-hour operating period was 57 wt.% based on maf coal plus hydrogen added. The yields of naphtha and light gas oil were 36 and 38 wt.% respectively.

A total of fourteen catalytic two-stage experiments were conducted using hydrogenated coal-derived oil as reaction solvent. Amocat 1C, a nickel-molybdenum catalyst with a bimodal pore size distribution, was used in both reactors. Experiments were carried out with Lingan Harbour, Prince Hub and Donkin Harbour seam coals to assess coal reactivity and the effects of first-stage reactor temperature, pressure and catalyst age on raw product yields. Illinois No. 6 coal was tested as a benchmark feedstock.

High coal conversions, 90 - 94 wt.%, were achieved in the catalytic two-stage processing mode. First-stage reactor temperature and catalyst age were found to have a greater influence on oil yield than pressure over the range of operating conditions investigated. At a first-stage temperature of 400°C and 17.24 MPa total pressure, oil yield from Lingan coal was 48 wt.% compared with 55 wt.% from Prince and Donkin and 59 wt.% from Illinois No. 6. All three Sydney area coals exhibited lower gas yield and higher levels of asphaltenes and preasphaltenes than the American coal.

Further work on catalytic two-stage liquefaction is planned. Immediate efforts will concentrate on catalyst and process design features aimed at reducing the level of asphaltenes in the raw product mix. In addition, process related options are being considered to maintain the quality of recycle solvent in coal liquefaction operations. The overall objective in the near term is to define the most promising two-stage process configuration for high rank coals from the Sydney coalfield.

## INTRODUCTION

In Canada, our way of life is highly dependent on petroleum liquids. Since our existing crude supplies are not expected to meet our future liquid fuel demands, it seems that we will have to rely on synthetic liquid fuels to some degree in the next few decades. Thus, should it be economically attractive or essential, abundant coal reserves are available in Canada for conversion to synthetic liquid fuels. Because long lead times are involved in process evaluation, detailed design and plant construction, it is essential to understand the basic concepts of the promising liquefaction processes and verify their potential yield structure as they apply to Canadian coals.

The in-situ coal resources of Nova Scotia occur in five main fields: Sydney, Mabou, Port Hood, Pictou and Springhill. The largest resources, over 2 billion metric tons, occur in the submarine area of the Sydney coalfield. The coal-bearing section of the Sydney basin contains eleven major seams ranging from 1 - 4.5 m thick. Historically, the two most productive seams have been the Harbour and Phalen seams.

At present, the principal coal producer in the province is the Cape Breton Development Corporation with two operating mines, Lingan and Prince, and a third under development at Donkin. All three mines are located in the Sydney area. The Harbour seam is currently being worked at Lingan whereas Hub seam coal is being produced from the Prince mine. Both coals are classified as high volatile A bituminous.

Process studies relating to the direct liquefaction of Nova Scotia high volatile bituminous coals were initiated at the Nova Scotia Research Foundation Corporation (NSRFC) in 1978. Initially, efforts were concentrated on a low-severity extractive coking process (1,2,3). From 1980 to 1984, experimental work was conducted on medium to high pressure donor extraction (4,5) and single-stage catalytic hydrogenation (6). Yield data has been developed over a wide range of operating conditions using bench-scale batch and continuous-flow hydroprocessing units. The product slate of interest is liquid transportation fuels.

Test results have shown that single-stage catalytic hydrogenation produces higher oil yields than donor solvent extraction. Furthermore, it has been shown that considerable control can be exercised over product distribution by varying process severity. Once-through experiments with Lingan Harbour seam coal and commercial nickel-molybdenum hydrotreating catalyst over medium to high severity operating conditions produced high coal conversions, 90 - 92%, while oil yields ranged from 30 - 47%.

Direct catalytic coal liquefaction processes, such as H-Coal, are single-stage processes. The catalyst serves several functions and the severe operating conditions required for one function might not be needed for other functions. Recent process developments in the United States are centered on liquefying coal in two separate reaction zones. Two-stage processing offers the versatility of controlling coal

dissolution and primary product upgrading under different operating conditions.

During the first half of 1985 our bench-scale coal liquefaction unit was modified and expanded for integrated two-stage operations. The closely coupled two reactor system offers the potential of higher distillate yields and improved hydrogen utilization. From mid-1985 to late 1986 the unit was operated with a first-stage thermal dissolver and a second-stage catalytic reactor. High coal conversions, 92 - 94%, were achieved in once-through experiments with Lingan Harbour, Donkin Harbour and Prince Hub seam coals using alumina base hydroprocessing catalysts in the second-stage reactor (7,8). In a six-day recycle liquefaction test with Prince coal and a nickel-molybdenum bimodal catalyst the net $C_4^+$ liquid yield was 67% during the last twelve-hour operating period. The yields of naphtha and light gas oil were 44 and 38 wt.% respectively (9).

Recently, an extension of the six-day recycle liquefaction test was undertaken to further investigate process yields and recycle solvent properties. On completion of that test the first-stage thermal dissolver was converted to a catalytic dissolver. Following this, a total of fourteen catalytic two-stage experiments were conducted to assess coal reactivity and the effects of first-stage reactor temperature, pressure and catalyst age on raw product distribution. Test results from the thermal/catalytic recycle operation and the catalytic two-stage experiments are presented in this report.

## TASK 1 - EQUIPMENT MAINTENANCE

Actual work on this task began in September of 1986. Activities included inspection and general maintenance of the hydrogen compression facility, the fixed-bed catalytic hydrotreater and the two-stage coal liquefaction unit.

In the hydrogen compression and storage area efforts were focused on the single-stage diaphragm compressor. The design capacity of this 7.5 horsepower unit is 114 L/min from 3.5 MPa to 34.5 MPa. Routine maintenance involved cleaning the process and hydraulic fluid filters, replacing the hydraulic fluid and checking the tension of the drive belts. Unscheduled maintenance was required late in October due to a diaphragm failure. After replacement of the triple diaphragm group and seals the compressor was returned to service.

The fixed-bed hydrotreater has been described in an earlier report (3). On turnaround, the high pressure liquid metering pump and the vacuum pump used regularly with this system were given a general overhaul. Liquid transfer lines between the feed vessel and the reactor inlet were removed and cleaned. Following this, new heating tapes and insulation were installed on these lines. In addition, high pressure valves on both feed and product vessels were replaced.

A complete description of the two-stage coal liquefaction unit is presented elsewhere (7). Most of the work associated with

Task 1 was related to the mechanical maintenance of this system. The first priority was to inspect the second-stage reactor internal components. On opening the reactor it was found that several parts of the agitator assembly were considerably worn. The lower drive shaft was replaced and a new bearing was installed in the base weldment. The reactor was then reassembled and returned to its position in the system. Several other parts on the rig were replaced including; all rupture discs, one thermocouple on the second-stage reactor, defective cooling water connections on magnedrive stirrers, worn high pressure block valves and frayed high temperature insulation. Worn product let-down valves were rebuilt as was the speed reducer on the Hills-McCanna slurry metering pump. Wet test meters were thoroughly cleaned and then performance tested.

The Fluke datalogger has been installed and is operational. It is presently equipped to log temperature and pressure inputs. Electrical components have been acquired recently to interface the hydrogen flow control system with the datalogger. These components along with external alarms activated by user defined alarm conditions will be added to the system over the next few months. Future plans include upgrading the feed weighing system to provide an output signal compatible with the datalogger.

## TASK 2 - PROCUREMENT OF NEW EQUIPMENT

The purpose of this task was to procure a 3.8-liter stirred tank reactor complete with Robinson/Mahoney style internals, furnace and temperature controller.

Two potential applications were envisaged for this reactor. Firstly, to replace the 1.5-liter first-stage reactor in order to conduct experiments over a wider range of slurry space velocity. Secondly, to function as a back-up for the installed second-stage reactor.

The reactor and internals purchased were manufactured by Autoclave Engineers Inc. of Erie, Pennsylvania. The internals consist of a catalyst basket assembly and an impeller/baffle system.

During operation, hot reacting slurry is constantly recirculated within the gradientless catalytic reactor. Slurry is forced through the annular basket and consequently across the catalyst by the impeller situated in the open area in the center of the basket. Slurry product and gases exist the reactor via an overflow pipe. This configuration permits catalyst to be used in bead or extrudate form.

## TASK 3 - PREPARATION OF RAW MATERIALS

### 3.1 Coal Feedstocks

Four coal feedstocks were tested in this study, three Sydney area coals and Illinois No. 6. Experimental work was concentrated on the Sydney area coals which are classified as high volatile bituminous in rank. Lingan Harbour, Donkin Harbour and Prince Hub seam coals were provided courtesy of the Cape Breton Development Corporation. Illinois No. 6 coal from the Burning Star Mine was procured through Cepheus Industries Incorporated, Marion, Illinois. Preparation involved riffling, grinding and screening. After screening, the individual coals were thoroughly mixed and stored in plastic-lined drums with a nitrogen blanket.

Proximate and ultimate analysis of the coals are given in Table 1. Proximate analysis, free-swelling index and forms of sulphur were determined according to standard methods. Total sulphur was measured using a Leco sulphur analyzer. Carbon, hydrogen and nitrogen were determined using a Perkin-Elmer elemental analyzer.

Petrographic analysis of the four coals is given in Table 2. These analyses were carried out by coal petrology experts at the Bedford Institute of Oceanography.

TABLE 1

ANALYSIS OF COALS TESTED

| COAL DESCRIPTION: | LINGAN HR-8 | DONKIN HR-8 | PRINCE HB-8 | ILLINOIS No.6 |
|---|---|---|---|---|
| Moisture, Wt. % | 1.02 | 1.83 | 3.22 | 3.03 |
| PROXIMATE (Dry Basis) Wt. % | | | | |
| Ash | 2.73 | 5.57 | 5.86 | 11.16 |
| Volatile Matter | 36.35 | 36.64 | 36.22 | 38.77 |
| ULTIMATE (Dry Basis) Wt. % | | | | |
| Carbon | 82.80 | 77.92 | 75.61 | 68.43 |
| Hydrogen | 5.40 | 5.00 | 5.09 | 4.71 |
| Nitrogen | 1.19 | 1.37 | 0.97 | 1.24 |
| Sulphur | 0.87 | 2.73 | 2.18 | 3.40 |
| Ash | 2.73 | 5.57 | 5.86 | 11.16 |
| Oxygen (by difference) | 7.01 | 7.41 | 10.35 | 11.06 |
| SULPHUR FORMS (Dry Basis) Wt. % | | | | |
| Sulphate | <0.01 | 0.20 | 0.06 | 0.31 |
| Pyritic | 0.47 | 1.26 | 1.49 | 1.09 |
| Organic (by difference) | 0.40 | 1.27 | 0.63 | 2.00 |
| Total | 0.87 | 2.73 | 2.18 | 3.40 |
| FREE SWELLING INDEX | 8½ | 7½ | 6½ | 1½ |
| HEATING VALUE (kj/kg) | 35,029 | 32,454 | 32,361 | 28,223 |

TABLE 2

PETROGRAPHIC CHARACTERISTICS OF COALS TESTED

| COAL DESCRIPTION: | LINGAN HR-8 | DONKIN HR-8 | PRINCE HB-8 | ILLINOIS No.6 |
|---|---|---|---|---|
| MACERAL COMPOSITION (% by Volume) | | | | |
| Vitrinite | 73.4 | 75.8 | 69.4 | 79.2 |
| Exinite | 5.4 | 3.2 | 5.2 | 3.8 |
| Inertinite | 20.8 | 16.8 | 23.6 | 11.8 |
| Macrinite | (2.2) | (1.4) | (5.2) | (0.6) |
| Micrinite | (6.0) | (7.8) | (8.4) | (4.6) |
| Semifusinite | (5.0) | (2.8) | (5.6) | (2.8) |
| Fusinite | (7.6) | (4.8) | (4.4) | (3.8) |
| Mineral Matter | 0.4 | 4.2 | 1.8 | 5.2 |
| Pyrite | -- | (2.6) | (0.4) | (1.4) |
| Quartz | -- | (0.4) | -- | (1.4) |
| Shale | -- | (1.2) | (1.4) | (2.4) |
| Calcite | -- | -- | -- | -- |
| % $R_0$ Max. | 1.00 | 0.92 | 0.79 | 0.62 |
| V-Type Distribution, % | | | | |
| V5 | -- | -- | -- | 32 |
| V6 | -- | -- | -- | 64 |
| V7 | -- | 2 | 58 | 4 |
| V8 | 6 | 36 | 34 | -- |
| V9 | 36 | 58 | 8 | -- |
| V10 | 58 | 4 | -- | -- |
| V11 | -- | -- | -- | -- |

## 3.2 Anthracene Oil

This material is a coal-tar distillate which contains ca. 5.8 wt.% hydrogen. It was supplied by Domtar Chemicals Limited (Hamilton, Ontario). Hydrogenated solvent used in the two-stage coal liquefaction experiments was made from two batches of this highly aromatic oil.

The characteristics of the raw oil are presented in Table 3. Carbon and hydrogen were determined using a Perkin-Elmer elemental analyzer. A standard Kjeldahl method was used for the determination of nitrogen. Total sulphur content was measured by x-ray fluorescence. Oxygen was determined by neutron activation.

In this study, the hydrogen aromatic/aliphatic distribution was established by proton nuclear magnetic resonance (H-NMR). A varian EM360L continuous wave spectrometer operating in the Fourier transform mode at 60 MHz, and a varian T60 NMR spectrometer operating in the Fourier transform mode at 60 MHz were used to obtain H-NMR spectra. Proton assignments were as follows: aromatic hydrogens ($H_{ar}$) were found in the region of 9 - 6 ppm. Benzylic hydrogens, including methyls, occur at 3.5 to 2 ppm ($H_{\alpha}$). Hydrogens, except methyls, on carbons one or more positions removed from the aromatic rings occur at 2 - 1.1 ppm ($H_{\beta}$). Aliphatic methyls are found at 1.1 - 0.4 ppm ($H_{\gamma}$). As almost no long aliphatic side chains are believed to be present, the $H_{\beta}$ protons are predominantly in rings. These assignments are consistent with similar analyses reported in the literature (10,11). Figure 1 is a typical H-NMR spectrum of raw anthracene oil.

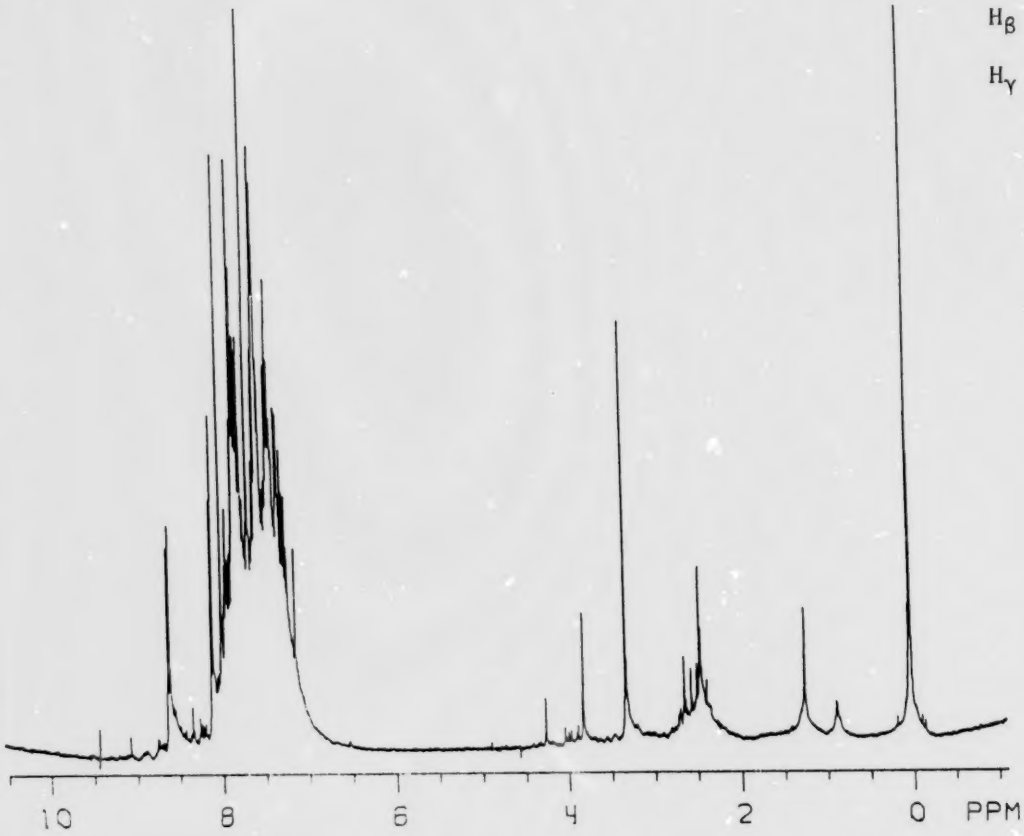
TABLE 3

ANALYSIS OF RAW ANTHRACENE OIL

| SAMPLE DESCRIPTION: | BATCH No. 7-16 | BATCH No. 7-17 |
|---|---|---|
| Specific Gravity, 60/60°F | 1.1347 | 1.1351 |
| Carbon, Wt. % | 90.46 | 90.97 |
| Hydrogen, Wt. % | 5.74 | 5.75 |
| Nitrogen, Wt. % | 0.84 | 0.89 |
| Sulphur, Wt. % | 0.55 | 0.55 |
| Oxygen, Wt. % | 1.00 | 0.97 |
| Proton Distribution, % | | |
| $H_{ar}$ (9 - 6 ppm) | 84.48 | 86.57 |
| $H_{\alpha}$ (3.5 - 2 ppm) | 9.39 | 8.57 |
| $H_{\beta}$ (2 - 1.1 ppm) | 3.25 | 3.14 |
| $H_{\gamma}$ (1.1 - 0.4 ppm) | 2.89 | 1.71 |
| Distillation Data °C (ASTM D-1160) | | |
| IBP | 205 | 213 |
| 10 % | 260 | 251 |
| 20 % | 291 | 299 |
| 30 % | 323 | 323 |
| 40 % | 339 | 336 |
| 50 % | 352 | 348 |
| 60 % | 365 | 358 |
| 70 % | 378 | 375 |
| 80 % | 395 | 397 |
| 90 % | 427 | 430 |
| 95 % | 450 | 451 |
| EP | 499 | 497 |

Figure 1 - H-NMR SPECTRA OF RAW ANTHRACENE OIL WITH 5.74 WT. % HYDROGEN

| PROTON DISTRIBUTION, | % |
|---|---|
| $H_{ar}$ (9 - 6 ppm) | 84.48 |
| $H_{\alpha}$ (3.5 - 2 ppm) | 9.39 |
| $H_{\beta}$ (2 - 1.1 ppm) | 3.25 |
| $H_{\gamma}$ (1.1 - 0.4 ppm) | 2.89 |



## 3.3 Catalysts

Two hydroprocessing catalysts were used in this study, Amocat 1C and Nalcomo 479. The former is an alumina-supported $NiO-MoO_3$ catalyst with a bimodal pore size distribution. It was used in the two-stage coal liquefaction experiments. Nalcomo 479 is an alumina-supported $CoO-MoO_3$ catalyst with a unimodal pore size distribution. It was used to prepare reaction solvent. Both catalysts were used in extrudate form. The chemical and physical properties of the two catalysts are given in Table 4.

TABLE 4

CHEMICAL AND PHYSICAL PROPERTIES OF HYDROTREATING CATALYSTS

| | AMOCAT 1C | NALCOMO 479 |
|---|---|---|
| $MoO_3$, Wt. % | 16.0 | 19.0 |
| NiO, Wt. % | 3.0 | - |
| CoO, Wt. % | - | 4.4 |
| Size, cm | 0.16 | 0.16 |
| Surface Area, $M^2/g$ | 190 | 180 |
| Pore Volume, cc/g | 0.85 | 0.47 |
| Density, g/cc | 0.67 | 0.82-0.84 |
| Crush Strength, kg | 4.4 | 6.4-7.3 |
| Modal Pore Diameter, A° | 100/1500 | 100 |

## TASK 4 - HYDROGENATION OF ANTHRACENE OIL

Reaction solvent used in Task 5 was prepared from raw anthracene oil using a fixed-bed catalytic hydrotreater. The bench-scale unit has been described in an earlier report (3).

A commercial cobalt-molybdenum hydrotreating catalyst, Nalcomo 479, was used to prepare solvent containing 9% hydrogen. A summary of the operating conditions used to produce the hydrogenated oil is given in Table 5. The characteristics of a 9%H oil blend are shown in Table 6. An H-NMR spectrum of 9%H solvent is presented in Figure 2. Hydrogenated anthracene oil, 7.5%H, spiked with dimethyl disulphide was used to sulphide the Amocat 1C catalyst prior to catalytic two-stage testing in Task 5. The spiked oil was on hand from previous sulphiding runs.

Approximately 300 liters of 9%H solvent was produced during this phase of work. The solvent was prepared at the rate of 1.8 liters per 7-hour shift. The catalytic hydrotreater was often operated two shifts per day. Daily production was collected to obtain 20-liter batches. Analysis of selected batches are presented in Table 7. About 45 liters of the hydrogenated oil was used for catalyst conditioning and post run operations. The analysis given in Table 6 is representative of a blend of the 20-liter batches.

TABLE 5

REACTION CONDITIONS FOR ANTHRACENE OIL HYDROGENATION

| | |
|---|---|
| Reactor Temperature, °C | 372-375 |
| Reactor Pressure, MPa | 15.51 |
| LHSV, $hr^{-1}$ | 0.20 |
| Hydrogen Flow, L/hr | 175-280 |
| Product Hydrogen Content, Wt. % | 9.0 |

Catalyst used: Nalcomo 479

TABLE 6

CHARACTERISTICS OF HYDROGENATED ANTHRACENE OIL

| SAMPLE DESCRIPTION: | HAO 81 |
|---|---|
| Specific Gravity, 60/60°F | 1.0285 |
| Carbon, Wt. % | 90.27 |
| Hydrogen, Wt. % | 9.02 |
| Nitrogen, Wt. % | 0.15 |
| Sulphur, Wt. % | 0.006 |
| Oxygen, Wt. % | 0.26 |
| Proton Distribution, % | |
| $H_{ar}$ (9 - 6 ppm) | 26.85 |
| $H_{\alpha}$ (3.5 - 2 ppm) | 29.23 |
| $H_{\beta}$ (2 - 1.1 ppm) | 34.67 |
| $H_{\gamma}$ (1.1 - 0.4 ppm) | 9.25 |
| Distillation Data, °C (ASTM D-1160) | |
| IBP | 185 |
| 10 % | 245 |
| 20 % | 267 |
| 30 % | 287 |
| 40 % | 301 |
| 50 % | 311 |
| 60 % | 321 |
| 70 % | 332 |
| 80 % | 349 |
| 90 % | 373 |
| 95 % | 390 |
| EP | 464 |

Figure 2 - H-NMR SPECTRA OF RAW ANTHRACENE OIL WITH 9.02 WT. % HYDROGEN

| PROTON DISTRIBUTION, % | |
|---|---|
| $H_{ar}$ (9 - 6 ppm) | 26.85 |
| $H_{\alpha}$ (3.5 - 2 ppm) | 29.23 |
| $H_{\beta}$ (2 - 1.1 ppm) | 34.67 |
| $H_{\gamma}$ (1.1 - 0.4 ppm) | 9.25 |

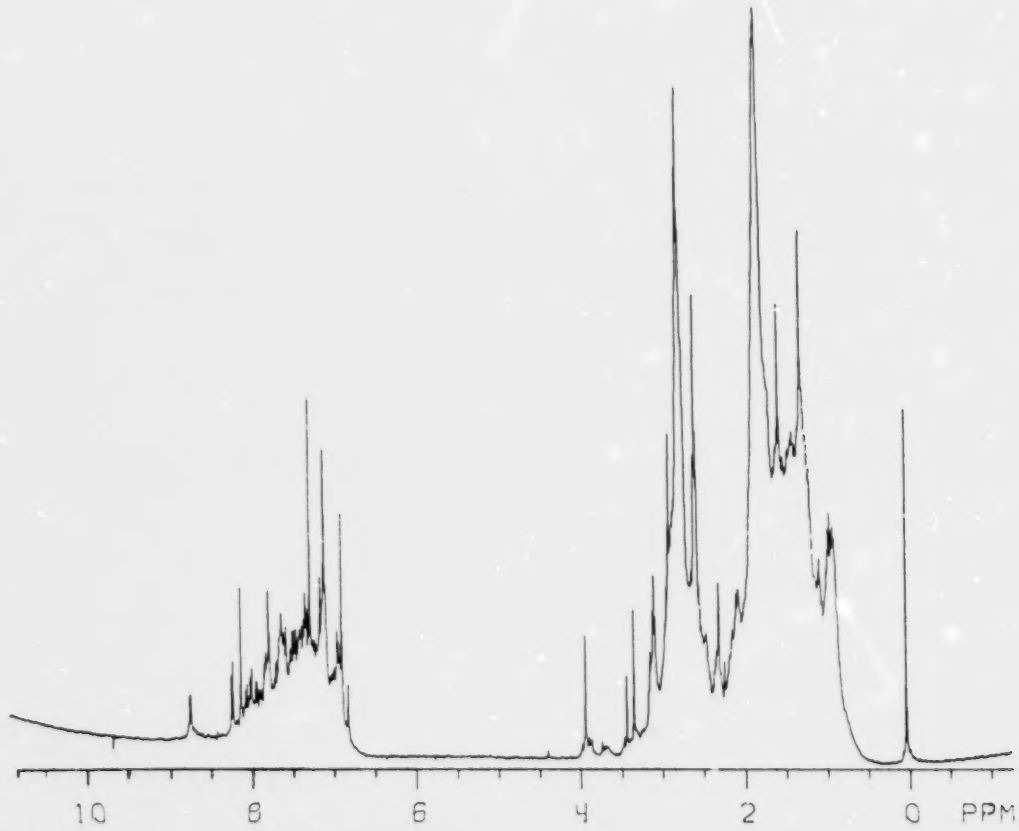


TABLE 7

ANALYSIS OF 20-LITER BATCHES OF 9% H SOLVENT

| BATCH NO. | 1 | 2 | 3 | 4 | 5 | 6 | 7 |
|---|---|---|---|---|---|---|---|
| Specific Gravity, 60/60°F | 1.0275 | 1.0275 | 1.0288 | 1.0281 | 1.0278 | 1.0258 | 1.0262 |
| Carbon, Wt. % | 90.05 | 90.20 | 89.70 | 89.83 | 90.02 | 89.75 | 89.68 |
| Hydrogen, Wt. % | 9.24 | 9.16 | 8.97 | 8.87 | 8.89 | 8.99 | 9.07 |
| Nitrogen, Wt. % | 0.14 | 0.15 | 0.15 | 0.15 | 0.14 | 0.14 | 0.13 |
| Sulphur, Wt. % | 0.006 | 0.006 | 0.006 | 0.008 | 0.006 | 0.006 | 0.007 |
| Proton Distribution, % | | | | | | | |
| $H_{ar}$ (9 - 6 ppm) | 26.33 | | | 27.33 | | | 25.08 |
| $H_{\alpha}$ (3.5 - 2 ppm) | 31.37 | | | 29.82 | | | 26.07 |
| $H_{\beta}$ (2 - 1.1 ppm) | 32.56 | | | 33.52 | | | 34.02 |
| $H_{\gamma}$ (1.1 - 0.4 ppm) | 9.74 | | | 9.33 | | | 14.84 |
| Distillation Data, °C (ASTM D-86) | | | | | | | |
| IBP | 190 | 180 | 175 | 194 | 181 | 194 | 196 |
| 5 % | 230 | 230 | 235 | 236 | 235 | 231 | 232 |
| 10 % | 246 | 251 | 255 | 253 | 260 | 251 | 250 |
| 20 % | 272 | 273 | 277 | 275 | 281 | 271 | 272 |
| 30 % | 288 | 287 | 293 | 292 | 295 | 287 | 290 |
| 40 % | 302 | 302 | 307 | 306 | 307 | 301 | 304 |
| 50 % | 313 | 315 | 319 | 319 | 318 | 314 | 315 |
| 60 % | 325 | 327 | 330 | 330 | 329 | 327 | 326 |
| 70 % | 336 | 339 | 342 | 342 | 340 | 338 | 338 |
| 80 % | 352 | 356 | 358 | 359 | 356 | 354 | 354 |
| 90 % | 376 | 380 | 383 | 387 | 381 | 380 | 378 |
| 95 % | 398 | 402 | 408 | 412 | 406 | 403 | 402 |
| EP | 525 | 502 | 511 | 508 | 506 | 497 | 506 |

TABLE 7 (Continued)

ANALYSIS OF 20-LITER BATCHES OF 9% H SOLVENT

| BATCH NO. | 8 | 9 | 10 | 11 | 12 | 13 | 14 |
|---|---|---|---|---|---|---|---|
| Specific Gravity, 60/60°F | 1.0295 | 1.0301 | 1.0288 | 1.0285 | 1.0295 | 1.0288 | 1.0275 |
| Carbon, Wt. % | 90.34 | 90.10 | 90.14 | 90.16 | 90.20 | 89.50 | 90.75 |
| Hydrogen, Wt. % | 8.94 | 8.96 | 8.90 | 8.95 | 8.83 | 9.01 | 9.13 |
| Nitrogen, Wt. % | 0.14 | 0.14 | 0.15 | 0.15 | 0.16 | 0.17 | 0.17 |
| Sulphur, Wt. % | 0.008 | 0.007 | 0.007 | 0.007 | 0.007 | 0.006 | 0.008 |
| Proton Distribution, % | | | | | | | |
| $H_{ar}$ (9 - 6 ppm) | | | 26.97 | | 28.02 | | 26.35 |
| $H_{\alpha}$ (3.5 - 2 ppm) | | | 28.86 | | 29.62 | | 29.87 |
| $H_{\beta}$ (2 - 1.1 ppm) | | | 34.80 | | 33.21 | | 34.77 |
| $H_{\gamma}$ (1.1 - 0.4 ppm) | | | 9.37 | | 9.16 | | 9.00 |
| Distillation Data, °C (ASTM D-86) | | | | | | | |
| IBP | 173 | 193 | 196 | 183 | 181 | 194 | 188 |
| 5 % | 230 | 235 | 230 | 231 | 235 | 230 | 228 |
| 10 % | 246 | 250 | 252 | 249 | 255 | 247 | 247 |
| 20 % | 271 | 274 | 272 | 273 | 275 | 270 | 270 |
| 30 % | 288 | 290 | 288 | 290 | 291 | 287 | 286 |
| 40 % | 303 | 304 | 302 | 303 | 305 | 301 | 300 |
| 50 % | 316 | 316 | 316 | 313 | 316 | 315 | 314 |
| 60 % | 331 | 328 | 328 | 326 | 327 | 328 | 326 |
| 70 % | 343 | 340 | 340 | 338 | 338 | 340 | 338 |
| 80 % | 360 | 355 | 355 | 353 | 353 | 356 | 354 |
| 90 % | 386 | 378 | 381 | 375 | 376 | 380 | 378 |
| 95 % | 414 | 399 | 408 | 396 | 394 | 403 | 402 |
| EP | 516 | 516 | 510 | 507 | 506 | 500 | 499 |

## TASK 5 - INTEGRATED TWO-STAGE LIQUEFACTION

In two-stage liquefaction processes, coal dissolution and upgrading reactions are conducted in two separate reaction zones. This approach permits the coal dissolution and catalytic upgrading steps to be independently optimized to maximize efficiency and control product distribution and quality.

The principal objectives of Task 5 were as follows: a) to further investigate recycle liquefaction of Prince Hub seam coal in a closely coupled two-stage thermal/catalytic process configuration; b) to initiate process studies on catalytic two-stage liquefaction applied to Sydney area coals.

A schematic diagram of the two-stage bench unit used is shown in Appendix G.

### 5.1 Thermal/Catalytic Testing With Liquid Recycle

During the previous phase of work a two-stage thermal/catalytic run of six days duration, RCL-7020, was conducted with liquid recycle using Prince Hub seam coal (9). On completion of RCL-7020 it was evident that the recycle liquid had not equilibrated. Hence, it was decided to extend the test by 6 - 7 days to further investigate recycle liquid properties, process yield and product quality. Following a shut-down period to procure more coal and perform routine equipment maintenance, run RCL-8000 was undertaken as an extension of RCL-7020.

A summary of the operating conditions for RCL-8000 is shown in Table 8. Catalyst age at the start-of-run was 261 kg coal/kg catalyst. The concentration of coal in the feed slurry was 28 wt.%. Hydrogen treatrate was fixed at 20 wt.% based on dry coal.

The operational configuration for this run is presented in Figure 3. Recycle liquid from RCL-7020 was used as start-up oil. Separator bottoms product was collected over 12-hour operating periods and then processed batchwise. Vacuum filtration was used to recover liquid from the separator bottoms. The recovered liquid was topped at 240°C by atmospheric distillation. Mid-heavy liquid in excess of the quantity required to mix with the feed coal was taken as net product.

Feed and product streams were analyzed to determine coal conversion, raw product distribution and hydrogen consumption. Samples of product slurry were extracted with tetrahydrofuran (THF) to establish coal conversion. Conversion was calculated on the basis of maf coal fed to the reaction zone over a 12-hour operating period and unconverted coal in the dry THF insoluble residue.

Vent gas samples were analyzed using a Perkin-Elmer Sigma 3B gas chromatograph interfaced with a Perkin-Elmer Sigma 10 chromatography data station. Water collected in the cold gas/liquid separator was analyzed for ammonia and sulphide using standard wet chemical methods. Hydrogen consumption was calculated on the basis of hydrogen fed and hydrogen measured in the vent gas streams.

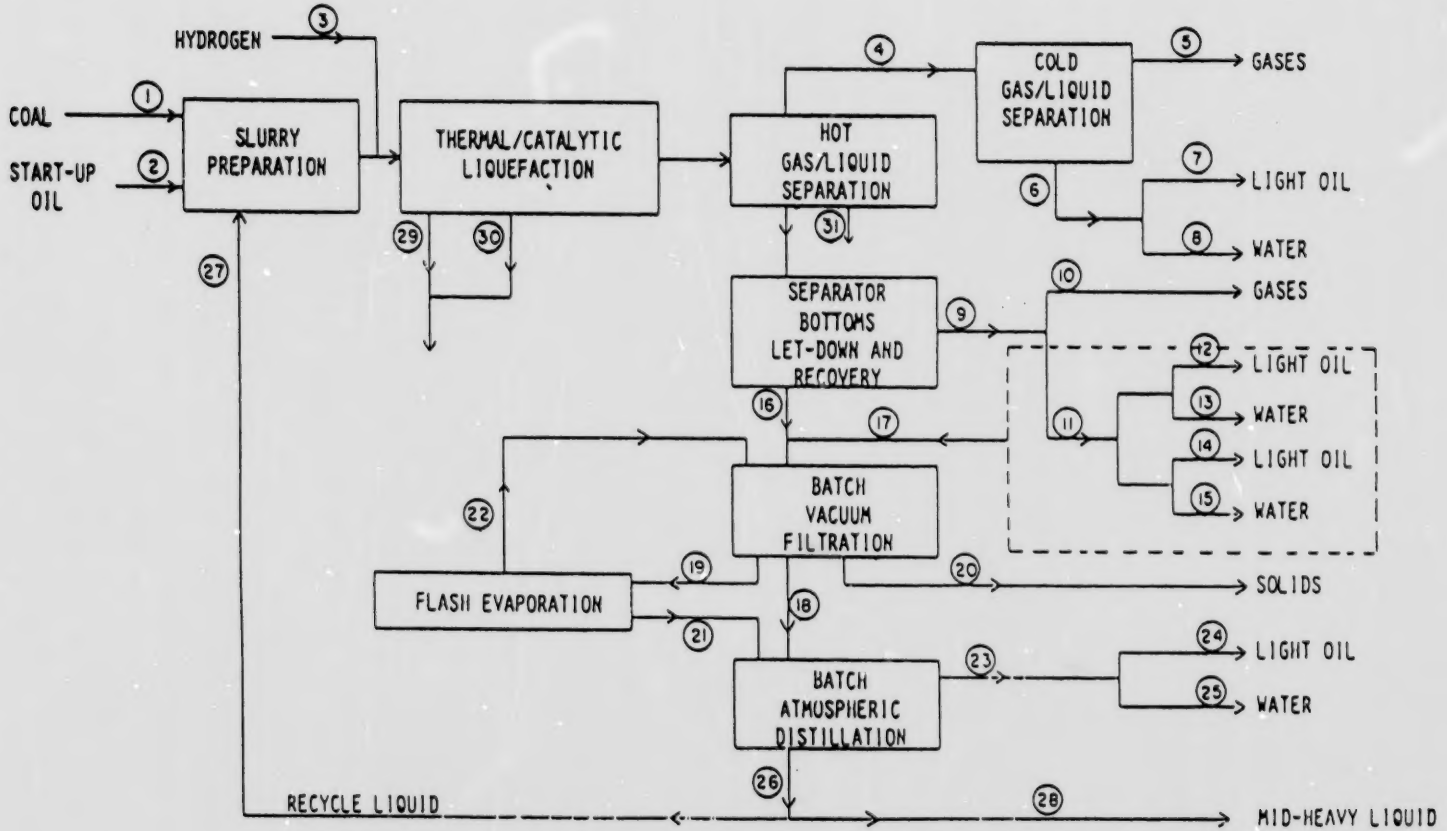
TABLE 8

SUMMARY OF OPERATING CONDITIONS FOR RCL-8000

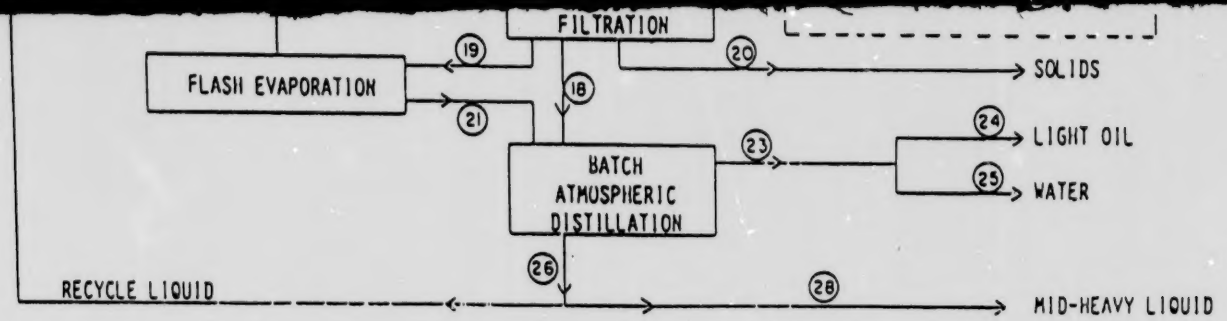
COAL: Prince Hub
CATALYST: Amocat 1C NiMo

| OPERATING CONDITIONS | HOURS ON STREAM | | | | | | |
|---|---|---|---|---|---|---|---|
| | 0 | 0 - 52 | 52 - 60 | 60 - 68 | 68 - 75 | 75 - 84 | 84 - 108 |
| Temperatures, °C: | | | | | | | |
| Thermal Dissolver | 440 | 440 | 450 | 455 | 455 | 455 | 450 |
| Catalytic Reactor | 435 | 435 | 435 | 440 | 445 | 440 | 435 |
| Back Pressure, MPa | 20.68 | 20.68 | 20.68 | 20.68 | 20.68 | 20.68 | 20.68 |
| SHSV, $hr^{-1}$ | | | | | | | |
| Thermal Dissolver | 1.00 | | | | | | |
| Catalytic Reactor | 0.33 | | | | | | |
| Coal Concentration, Wt. % | 28 | 28 | 28 | 28 | 28 | 28 | 28 |
| $H_2$ Treatrate, Wt. % Dry Coal | 20 | 20 | 20 | 20 | 20 | 20 | 20 |

Figure 3 - OPERATIONAL CONFIGURATION FOR RCL-8000




| STREAM NO. | DESCRIPTION |
|---|---|
| 1 | Coal |




| STREAM NO. | DESCRIPTION |
|---|---|
| 1 | Coal |
| 2 | Start-Up Oil |
| 3 | Hydrogen |
| 4 | Hot Separator Overheads |
| 5 | Primary Vent Gas |
| 6 | High Pressure Knock-Out Bottoms |
| 7 | High Pressure Knock-Out - Light Oil |
| 8 | High Pressure Knock-Out - Water |
| 9 | Low Pressure Product Receiver Overheads |
| 10 | Secondary Vent Gas |
| 11 | Low Pressure Product Receiver Overhead Condensate |
| 12 | Separator Bottoms Flash Knock-Out No. 1 - Light Oil |
| 13 | Separator Bottoms Flash Knock-Out No. 1 - Water |
| 14 | Separator Bottoms Flash Knock-Out No. 2 - Light Oil |
| 15 | Separator Bottoms Flash Knock-Out No. 2 - Water |
| 16 | Flashed Separator Bottoms |
| 17 | Condensate from SBFKO No. 1 and No. 2 |
| 18 | Filtered Separator Bottoms |
| 19 | Cyclohexane Wash |
| 20 | Washed Filter Cake |
| 21 | Cyclohexane Solubles Recovered from Filter Cake |
| 22 | Recycled Cyclohexane |
| 23 | Atmospheric Still Overheads |
| 24 | Atmospheric Still Light Oil |
| 25 | Atmospheric Still Water |
| 26 | Atmospheric Still Bottoms |
| 27 | Recycle Liquid |
| 28 | Mid-Heavy Liquid Product |
| 29 | Thermal Dissolver Contents |
| 30 | Catalytic Reactor Contents |
| 31 | Separator No. 1 Contents |

Run plots showing coal conversion, $C_1$ - $C_3$ gas production, total gas make and hydrogen consumption are shown in Figure 4. A summary of performance and material balance data for each 12-hour operating period is given in Table 9. Detailed information is provided in Appendices B and C.

Physical and chemical characteristics of the recycle liquid were monitored throughout RCL-8000. Run plots in Figures 5 and 6 show the response of recycle liquid properties as a function of time. Hydrogen aromatic/aliphatic distributions are presented in Table 10.

Coal conversion, gas make and hydrogen consumption for the first 12-hour operating period of RCL-8000 were 92.9, 30.7 and 7.2%. These levels were similar to those of the last 12-hour operating period of RCL-7020. However, by the end of the second day hydrogen consumption had decreased to 6.1%. Furthermore, the specific gravity of the recycle liquid increased steadily from 1.0605 to 1.0734 during the first two days of operation as the pentane insolubles rose from 13.1 to 18.2%. As a direct result of this the filtration rate of separator bottoms product slowed considerably. At this point there was concern that if these trends continued it would soon be impossible to maintain solvent balance.

In an effort to promote thermal cracking in the first-stage, the temperature of the dissolver was increased from 440 to 450°C at hour 52. It was increased further to 455°C at hour 60 and

held at that temperature for 24 hours. The catalytic reactor was operated at 435°C from 0 to 60 hours. From hour 60 to hour 84 it was operated at 440 - 445°C in an effort to promote liquid phase hydrogenation and hydrocracking reactions. These changes in the first and second-stage reactor temperatures did not produce the desired effects. During this period coal conversion decreased to 89.9% and the specific gravity of the recycle liquid increased to 1.0914. In addition, gas make rose substantially to 37.1%. At hour 84 the thermal dissolver and catalytic reactor temperatures were lowered to 450 and 435°C respectively to decrease gas production.

The data presented in Table 9 illustrates that gas production declined after hour 84 in response to lowering reactor temperatures. Unfortunately, coal conversion and hydrogen consumption also declined. Figures 5 and 6 illustrate that specific gravity, pentane insolubles, nitrogen content and sulphur content of the recycle liquid increased steadily as the run progressed whereas hydrogen content decreased. The direction and the extent of these trends are indicative of significant catalyst fouling. Hence, RCL-8000 was terminated at hour 108.

Performance data and net liquid product properties for the last 12-hour operating period are shown in Table 11. Coal conversion over the 96 - 108 hour period averaged 85.0%. The net $C_4^+$ liquid yield was 57.2%. Inspections were carried out on the $C_5^+$ liquid product. This liquid had a specific gravity of 0.9826 and was high in pentane

insolubles, 10.26%. The distillation data shows 80 vol % of the liquid to be light and middle oil distillate at less than 375°C.

At hour 108 a test procedure was undertaken to assess the condition of the Amocat 1C catalyst. On the assumption that catalyst fouling had occurred, the objective was to determine if it was reversible. The test procedure consisted of two steps. First, a 48 hour flush operation designed to remove adsorbed asphaltenes from the catalyst. Hydrogenated anthracene oil spiked with dimethyl disulphide was used as a flush solvent. Second, a once-through coal liquefaction experiment with a previously tested coal/oil feedstock to measure catalyst performance. On completion of the second step the unit was flushed and then shut-down after 8 consecutive days of operation.

Experimental results for the once-through liquefaction experiment, Run No. 8001, are presented below:

| Description | Run No. 7018 | Run No. 8001 |
|---|---|---|
| Catalyst Age, kg coal/kg catalyst | 112 | 343 |
| Coal Conversion, Wt. % | 94.5 | 91.6 |
| Product Distribution, Wt. % maf Coal | | |
| Gases | 31.1 | 32.6 |
| Oil | 48.5 | 26.5 |
| Asphaltenes | 12.6 | 23.5 |
| Preasphaltenes | 2.3 | 9.0 |
| Unreacted Coal | 5.5 | 8.4 |
| Hydrogen Consumption, Wt. % maf Coal | 7.6 | 5.9 |

The results confirm that severe catalyst deactivation had occurred since Run No. 7018. Furthermore, the catalyst was not regenerated by washing with anthracene oil.

A few days after the unit was shut-down it was empirically established that the liquid phase capacity of the second-stage reactor at NTP was markedly less than design. Hence, the reactor was removed from the system and opened. A substantial accumulation of coke was found in the vessel. A solid mass of coke had formed between the bottom cover of the reactor and the base weldment and between the reactor wall and the catalyst basket. The coke build-up between the reactor wall and the basket extended into the catalyst bed.

Removal of the reactor internals proved to be a difficult task. It was necessary to drill out most of the coke to free the catalyst basket assembly. The reactor and the basket were both damaged during this procedure. Catalyst and coke were removed from the basket by a combination of drilling and heat treatment. The body of the reactor and the catalyst basket will have to be refurbished prior to further use.

In two-stage thermal/catalytic liquefaction it is essential to maintain recycle solvent quality. Data presented in Table 10, in particular hydrogen content and hydrogen proton distribution, indicate that the donor capacity of the recycle liquid decreased as run RCL-8000 progressed. As solvent quality decreased; coal conversion decreased, liquid product yield fell off and product quality declined. Furthermore, as the concentration of heavy asphaltenic materials in the recycle liquid increased so did its propensity to coke. This resulted in extensive coke formation in the second-stage catalytic reactor which was operating at high temperature and low slurry space velocity.

Figure 4 - RUN PLOTS OF COAL CONVERSION, $C_1$ - $C_3$ GAS PRODUCTION, TOTAL GAS MAKE AND HYDROGEN CONSUMPTION FOR RCL-8000

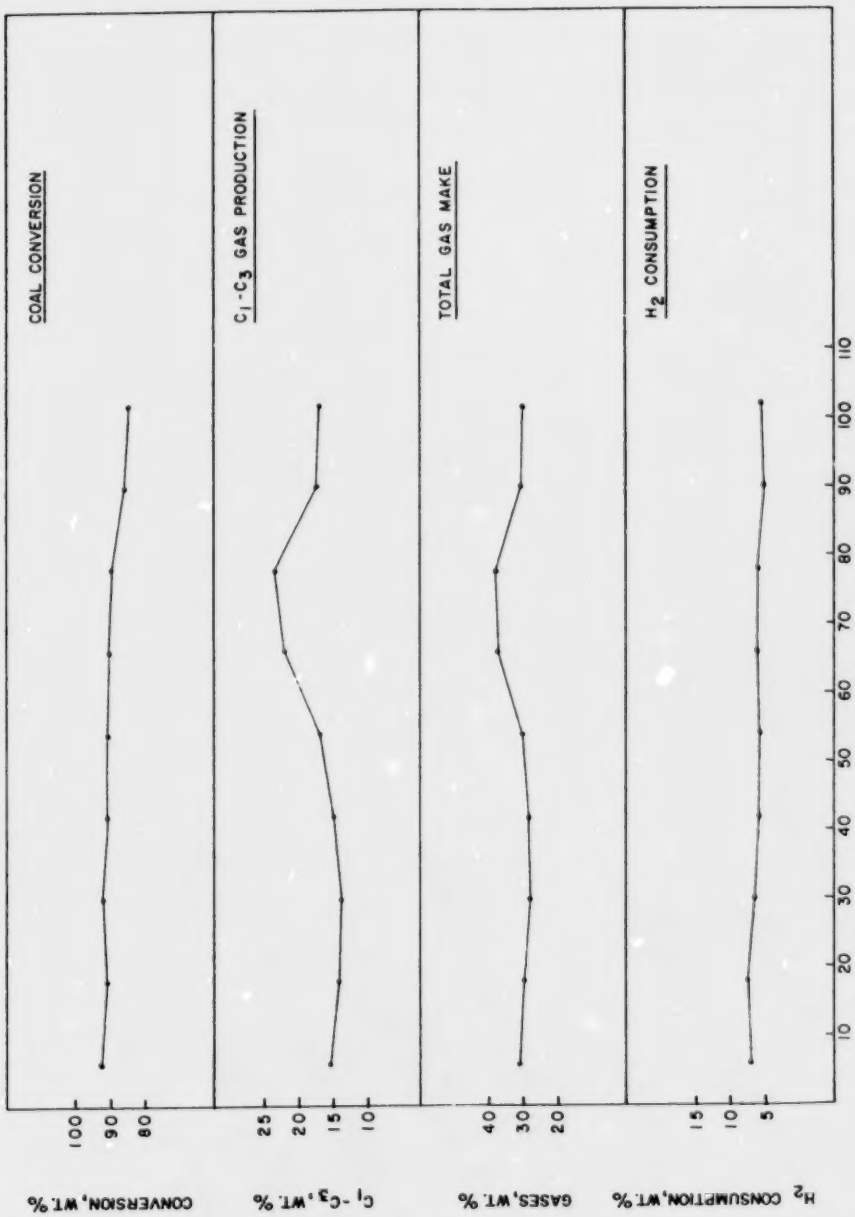


TABLE 9

PERFORMANCE AND MATERIAL BALANCE DATA FOR RCL-8000

| Operating Period (hrs.) | Coal Conversion (Wt. % maf coal) | $C_1 - C_3$ (Wt. % maf coal) | Total Gases (Wt. % maf coal) | Hydrogen Consumption (Wt. % maf coal) | Material Balance Liquefaction Step (Wt. %) |
|---|---|---|---|---|---|
| 0 - 12 | 92.9 | 15.3 | 30.7 | 7.2 | 100.5 |
| 12 - 24 | 91.6 | 14.1 | 29.5 | 7.4 | 98.6 |
| 24 - 36 | 92.6 | 14.1 | 28.0 | 6.5 | 98.1 |
| 36 - 48 | 91.3 | 15.0 | 28.4 | 6.1 | 97.1 |
| 48 - 60 | 91.5 | 17.1 | 30.5 | 6.1 | 97.0 |
| 60 - 72 | 90.6 | 22.3 | 37.1 | 6.6 | 96.2 |
| 72 - 84 | 89.9 | 23.7 | 37.9 | 6.1 | 96.3 |
| 84 - 96 | 86.3 | 17.5 | 30.6 | 5.7 | 96.5 |
| 96 - 108 | 85.0 | 17.1 | 30.1 | 5.8 | 101.6 |

Figure 5 - RESPONSE OF RECYCLE LIQUID SPECIFIC GRAVITY, HYDROGEN CONTENT AND PENTANE INSOLUBLES IN RCL-8000

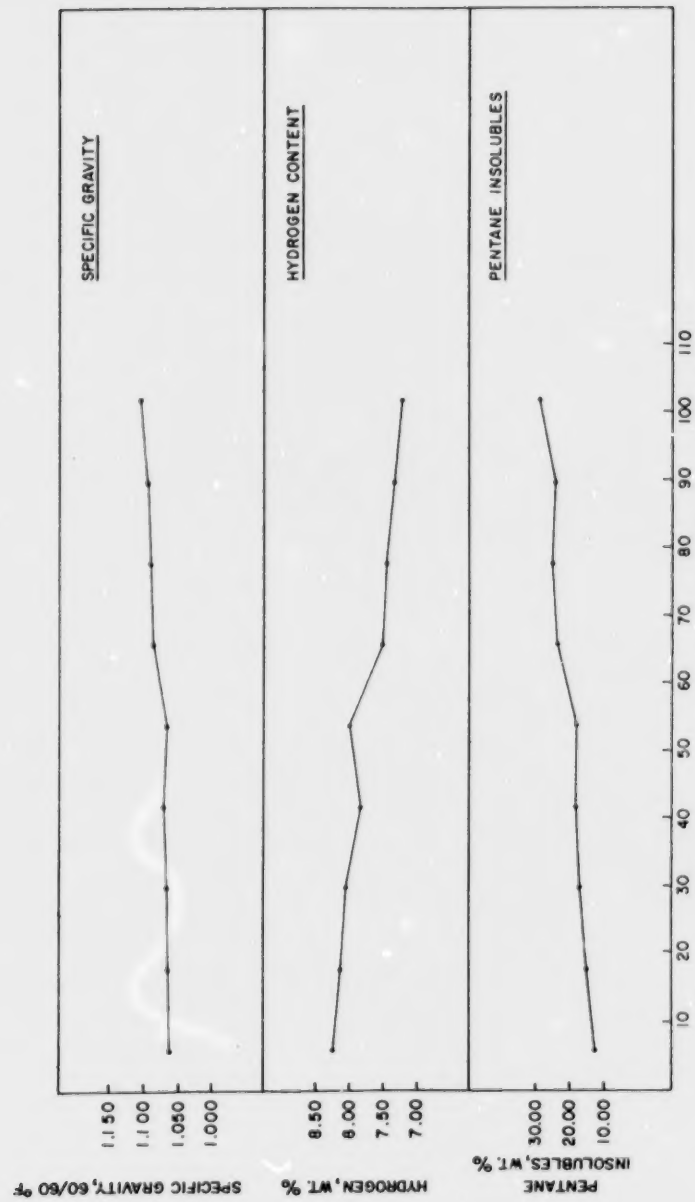


Figure 6 - RESPONSE OF RECYCLE LIQUID NITROGEN CONTENT AND SULPHUR CONTENT IN RCL-8000

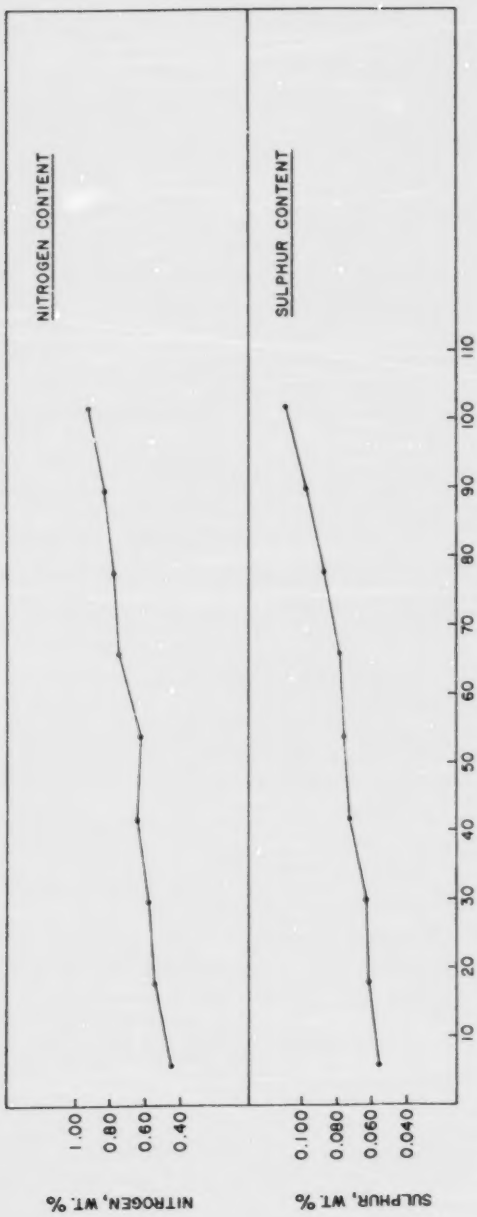


TABLE 10

PROPERTIES OF RECYCLE LIQUID FROM RCL-8000

| OPERATING PERIOD, HRS. | 0 - 12 | 12 - 24 | 24 - 36 | 36 - 48 | 48 - 60 | 60 - 72 | 72 - 84 | 84 - 96 | 96 - 108 |
|---|---|---|---|---|---|---|---|---|---|
| Specific Gravity, 60/60°F | 1.0605 | 1.0645 | 1.0675 | 1.0734 | 1.0698 | 1.0884 | 1.0914 | 1.0957 | 1.1054 |
| Carbon, Wt. % | 88.64 | 89.91 | 88.23 | 90.18 | 88.40 | 89.57 | 88.59 | 89.81 | 87.74 |
| Hydrogen, Wt. % | 8.27 | 8.18 | 8.09 | 7.88 | 8.02 | 7.52 | 7.49 | 7.38 | 7.23 |
| Nitrogen, Wt. % | 0.45 | 0.55 | 0.58 | 0.64 | 0.65 | 0.75 | 0.79 | 0.84 | 0.93 |
| Sulphur, Wt. % | 0.056 | 0.062 | 0.064 | 0.073 | 0.077 | 0.079 | 0.088 | 0.098 | 0.111 |
| Pentane Insolubles, Wt. % | 13.10 | 15.48 | 16.95 | 18.23 | 18.17 | 23.70 | 25.23 | 24.93 | 29.10 |
| Proton Distribution, % | | | | | | | | | |
| $H_{ar}$ (9-6 ppm) | 31.63 | | 32.13 | | 35.66 | | 39.31 | | 42.58 |
| $H_{\alpha}$ (3.5-2 ppm) | 27.38 | | 26.29 | | 25.84 | | 28.15 | | 26.06 |
| $H_{\beta}$ (2-1.1 ppm) | 27.99 | | 26.87 | | 27.00 | | 22.58 | | 20.83 |
| $H_{\gamma}$ (1.1-0.4 ppm) | 13.00 | | 14.70 | | 11.50 | | 9.96 | | 10.53 |
| Distillation Data, Wt. %: (Spinning Band) | | | | | | | | | |
| IBP - 205°C | | | | | | | | | 2.3 |
| 205 - 335 | | | | | | | | | 38.0 |
| 335 - 415 | | | | | | | | | 19.8 |
| 415 - 431°C | | | | | | | | | 3.2 |
| RESID | | | | | | | | | 36.7 |

TABLE 11

PERFORMANCE DATA AND NET LIQUID PRODUCT PROPERTIES FOR RCL-8000

| | |
|---|---|
| Coal Conversion, Wt. % | 85.0 |
| Gas Make, Wt. % | 30.1 |
| $C_4^+$ Liquid Yield, Wt. % | 57.2 |
| $C_5^+$ Liquid Yield, Wt. % | 53.9 |
| $H_2$ Consumption, Wt. % | 5.8 |
| $C_5^+$ Liquid Product Inspections: | |
| Specific Gravity, 60/60°F | 0.9826 |
| Carbon, Wt. % | 89.18 |
| Hydrogen, Wt. % | 8.60 |
| Nitrogen, Wt. % | 0.60 |
| Oxygen, Wt. % | 1.31 |
| Sulphur, Wt. % | 0.100 |
| Pentane Insolubles, Wt. % | 10.26 |
| Distillation Data, °C: (ASTM D-1160) | |
| IBP | 44 |
| 10% V/20 | 116/147 |
| 30/40 | 196/215 |
| 50/60 | 231/250 |
| 70/80 | 297/373 |
| 90/95 | - / - |
| EP | 515 (86% V) |
| Distillation Data, Wt. %: (Spinning Band) | |
| IBP - 205°C | 36.3 |
| 205 - 335 | 38.4 |
| 335 - 420°C | 9.3 |
| RESID | 16.0 |

Since RCL-8000 was an extension of RCL-7020 the recycle liquefaction operation with Prince coal was actually of 10½ days duration. Coal conversion, yield data and liquid product properties on day 6 (RCL-7020) and day 11 (RCL-8000) are shown in Table 12. The decrease in performance from day 6 to day 11 is primarily attributed to the coking that occurred after day 6. However, it is likely that process performance would have declined to some degree as the catalyst aged even if extensive coking had not occurred.

One approach to sustaining a constant liquid yield in commercial practice would be to maintain catalyst activity at a specific level by using a catalyst addition/withdrawal system. Using this procedure, liquid yield could be controlled as a function of catalyst consumption. Ideally, spent catalyst could be regenerated and used for more than one cycle. Furthermore, it is expected that catalyst activity could be maintained more effectively by controlling the level of asphaltenic material in the liquid recycle stream. However, this would be accomplished at the expense of liquid yield and thus a trade-off would have to be made between liquid yield and catalyst consumption. In conclusion, recycle liquefaction test work completed to date illustrates that more effort is required to define a process for two-stage thermal/catalytic liquefaction of Sydney area coals.

TABLE 12

COMPARISON OF YIELDS FROM TWO-STAGE THERMAL/CATALYTIC RECYCLE LIQUEFACTION OF PRINCE HUB SEAM COAL

| | RCI-7020 | RCL-8000 |
|---|---|---|
| Operating Period, hrs. | 132-144 | 96-108 |
| Accumulated On-Stream Time, hrs. | 132-144 | 240-252 |
| Coal Conversion, Wt. % | 91.3 | 85.0 |
| Gas Make, Wt. % | 32.0 | 30.1 |
| $C_4^+$ Liquid Yield, Wt. % | 67.0 | 57.2 |
| $C_5^+$ Liquid Yield, Wt. % | 62.8 | 53.9 |
| $H_2$ Consumption, Wt. % | 7.2 | 5.8 |
| $C_5^+$ Liquid Product Inspections: | | |
| Specific Gravity, 60/60°F | 0.8976 | 0.9826 |
| Carbon, Wt. % | 88.21 | 89.18 |
| Hydrogen, Wt. % | 10.69 | 8.60 |
| Nitrogen, Wt. % | 0.25 | 0.60 |
| Oxygen, Wt. % | 0.78 | 1.31 |
| Sulphur, Wt. % | 0.071 | 0.100 |
| Pentane Insolubles, Wt. % | 3.01 | 10.26 |
| Distillation Data, °C: (ASTM D-1160) | | |
| IBP | 47 | 44 |
| 10% V/20 | 89/102 | 116/147 |
| 30/40 | 122/174 | 196/215 |
| 50/60 | 206/226 | 231/250 |
| 70/80 | 236/283 | 297/373 |
| 90/95 | 372/ - | - / - |
| EP | 523 (94% V) | 515 (86% V) |

## 5.2 Catalytic Two-Stage Testing

In two-stage liquefaction a catalytic first-stage reactor offers potential for lower severity coal dissolution than a thermal dissolver. The presence of an effective hydrogenation catalyst in the first-stage would enhance the donor capacity of the recycle solvent and contribute to the stabilization of the initial products of coal dissolution. On completion of Task 5.1 the thermal dissolver of our bench unit was converted to a catalytic reactor to assess the potential of catalytic two-stage liquefaction applied to Nova Scotia coals.

The thermal dissolver was converted to a catalytic reactor by installing a catalyst basket assembly and a new impeller/baffle system in the vessel. The internal components were supplied by Autoclave Engineers Incorporated. However, a new support for the base weldment had to be fabricated at NSRFC's machine shop and broken welds on the catalyst basket also had to be repaired at NSRFC. Before reassembling the reactor the catalyst basket was charged with 0.1 kg of Amocat 1C nickel-molybdenum catalyst.

As mentioned earlier the second-stage reactor and catalyst basket used in RCL-8000 were damaged while drilling coke from the vessel after the test. In order to minimize downtime it was decided to use the new 3.8-liter reactor system as a replacement for the damaged one. After charging the catalyst basket with 0.3 kg of Amocat 1C the reactor was reassembled. Unfortunately, the new reactor had to be

assembled four times before it was put into service due to gas leaks detected during pressure tests with hydrogen at room temperature.

After the modified 1.5-liter first-stage reactor and the new 3.8-liter second-stage reactor were installed, the coal liquefaction unit was pressure tested at room temperature. Following this a trial run was made with oil alone to verify the operational status of mechanical and electrical systems.

Prior to initiating coal liquefaction experiments, a 30-hour run was carried out to sulphide the fresh catalyst using hydrogenated anthracene oil, 7.5%H, spiked with dimethyl disulphide. The procedure was conducted at 17.24 MPa and a start-of-run temperature of 175°C. Temperature was increased gradually over a 24-hour period to a maximum of 370°C.

A total of fourteen catalytic two-stage liquefaction experiments were completed under Task 5.2. Test work was concentrated on Harbour and Hub seam coals from the Sydney coalfield. One run was carried out with Illinois No. 6 coal for comparative purposes. The experiments were conducted in a once-through processing mode using hydrogenated anthracene oil containing 9%H as reaction solvent. Coal concentration in the feed slurry was fixed at 28 wt.%. Hydrogen treatrate was ca. 20 wt.% based on dry coal.

A typical run was of 10 hours duration. However, an additional 8 hours was required for preliminary and post run operations. In the majority of runs the controlled variables lined-out about 4 - 5 hours after start-up. During the steady state operating period vent gas streams were sampled and other product streams were collected for analytical and material balance purposes.

In these experiments the following definitions for the product fractions isolated by solvent extraction were used:

Oil - pentane solubles

Asphaltenes - toluene solubles, pentane insolubles

Preasphaltenes - THF solubles, toluene insolubles

A direct extraction technique was used to determine these fractions. Individual samples of product slurry were separately extracted with pentane, toluene and tetrahydrofuran (THF). Conversions were calculated on the basis of maf coal fed to the reaction zone during the steady state sampling period and unconverted coal in the dry insoluble residues. Conversion calculations are described in Appendix A.

The first experiment, Run No. 8002, was made to condition the catalyst. This 24-hour run was conducted with Lingan Harbour seam coal at a slurry feedrate of 1.5 kg/hr and a total pressure of 17.24 MPa. First and second-stage reactor temperatures were 400 and 425°C respectively. A comparison of results from Run Nos. 8002 and

7007 are shown below. Run No. 7007 was the catalyst conditioning operation carried out earlier at thermal/catalytic processing conditions.

| | Run No. 8002 | Run No. 7007 |
|---|---|---|
| Coal Conversion, Wt. % | 90.5 | 91.5 |
| Product Distribution, Wt. % maf Coal | | |
| Gases | 13.0 | 19.2 |
| Oil | 44.1 | 42.4 |
| Asphaltenes | 20.7 | 23.1 |
| Preasphaltenes | 12.7 | 6.8 |
| Unreacted Coal | 9.5 | 8.5 |
| Hydrogen Consumption, Wt. % maf Coal | 5.2 | 4.6 |

Operating conditions for Run No. 7007 were similar to those of Run No. 8002 except the first-stage dissolver was operated at 450°C in Run No. 7007. The catalytic two-stage run exhibited lower gas make, higher oil yield and a higher level of preasphaltenes in the raw product mix than the two-stage thermal/catalytic run. As expected, hydrogen consumption was higher in the catalytic two-stage processing mode.

After completion of the catalyst conditioning operation a series of experiments were carried out to investigate coal reactivity and the effects of first-stage reactor temperature, pressure, stirring rate and catalyst age on raw product yields. A summary of the results from these experiments is shown in Table 13. Detailed performance and material balance data are presented in Appendices B and C.

TABLE 13

RESULTS FROM CATALYTIC TWO-STAGE LIQUEFACTION EXPERIMENTS

| RUN NO.<br>COAL TESTED | 8002<br>LINGAN HR-6 | 8003<br>PRINCE HB-8 | 8004<br>DONKIN HR-8 | 8005<br>LINGAN HR-8 | 8006<br>LINGAN HR-8 | 8007<br>DONKIN HR-8 | 8008<br>PRINCE HB-8 |
|---|---|---|---|---|---|---|---|
| Temperatures: °C | | | | | | | |
| First-Stage Reactor | 415 | 400 | 400 | 400 | 420 | 420 | 420 |
| Second-Stage Reactor | 425 | 425 | 425 | 425 | 425 | 425 | 425 |
| Back Pressure, MPa | 17.24 | 17.24 | 17.24 | 17.24 | 17.24 | 17.24 | 17.24 |
| Slurry Feedrate, kg/hr | 1.546 | 1.006 | 1.041 | 1.048 | 1.037 | 1.019 | 1.028 |
| Hydrogen Treatrate, Wt. % Dry Coal | 9.7 | 19.8 | 19.3 | 19.1 | 19.4 | 19.7 | 19.8 |
| Catalyst Age, kg coal/kg catalyst | | | | | | | |
| First-Stage Reactor | 107 | 144 | 175 | 206 | 237 | 268 | 299 |
| Second-Stage Reactor | 31 | 42 | 51 | 60 | 69 | 78 | 87 |
| CONVERSIONS, WT. % OF maf COAL | | | | | | | |
| Pentane Solubles plus Gases | 57.1 | 75.0 | 73.5 | 61.8 | 67.4 | 77.3 | 78.4 |
| Toluene Solubles plus Gases | 77.8 | 87.3 | 87.2 | 78.9 | 83.6 | 89.4 | 88.8 |
| THF Solubles plus Gases | 90.5 | 92.2 | 92.9 | 90.0 | 91.1 | 93.7 | 92.8 |
| PRODUCT DISTRIBUTION, WT. % OF maf COAL | | | | | | | |
| Gases | 13.0 | 19.6 | 18.8 | 14.1 | 16.3 | 19.6 | 21.5 |
| Oil | 44.1 | 55.4 | 54.7 | 47.7 | 51.1 | 57.7 | 56.9 |
| Asphaltenes | 20.7 | 12.3 | 13.7 | 17.1 | 16.2 | 12.1 | 10.4 |
| Preasphaltenes | 12.7 | 4.9 | 5.7 | 11.1 | 7.5 | 4.3 | 4.0 |
| $H_2$ CONSUMPTION, WT. % OF maf COAL | 5.2 | 8.3 | 7.6 | 6.6 | 7.5 | 8.7 | 8.6 |

TABLE 13 (Continued)

RESULTS FROM CATALYTIC TWO-STAGE LIQUEFACTION EXPERIMENTS

| RUN NO.<br>COAL TESTED | 8009<br>PRINCE HB-8 | 8010<br>DONKIN HR-8 | 8011<br>ILLINOIS NO. 6 | 8012<br>DONKIN HR-8 | 8014<br>DONKIN HR-8 | 8015<br>DONKIN HR-8 | 8016<br>DONKIN HR-8 |
|---|---|---|---|---|---|---|---|
| Temperatures: °C | | | | | | | |
| First-Stage Reactor | 420 | 400 | 400 | 400 | 440 | 420 | 420 |
| Second-Stage Reactor | 425 | 425 | 425 | 425 | 425 | 425 | 425 |
| Back Pressure, MPa | 17.24 | 20.68 | 17.24 | 17.24 | 17.24 | 17.24 | 17.24 |
| Slurry Feedrate, kg/hr | 1.000 | 1.028 | 1.040 | 1.049 | 1.032 | 0.999 | 1.000 |
| Hydrogen Treatrate, Wt. % Dry Coal | 20.3 | 19.5 | 19.3 | 19.1 | 19.6 | 20.1 | 20.0 |
| Catalyst Age, kg coal/kg catalyst | | | | | | | |
| First-Stage Reactor | 330 | 361 | 392 | 423 | 454 | 486 | 518 |
| Second-Stage Reactor | 96 | 105 | 114 | 123 | 132 | 141 | 150 |
| CONVERSIONS, WT. % OF maf COAL | | | | | | | |
| Pentane Solubles plus Gases | 77.2 | 72.5 | 84.1 | 66.3 | 76.5 | 69.4 | 69.1 |
| Toluene Solubles plus Gases | 87.8 | 86.4 | 91.3 | 81.6 | 89.4 | 84.9 | 85.2 |
| THF Solubles plus Gases | 92.3 | 93.3 | 94.3 | 92.9 | 93.6 | 93.0 | 93.1 |
| PRODUCT DISTRIBUTION, WT. % OF maf COAL | | | | | | | |
| Gases | 21.1 | 20.4 | 25.0 | 16.5 | 23.3 | 19.9 | 20.6 |
| Oil | 56.1 | 52.1 | 59.1 | 49.8 | 53.2 | 49.5 | 48.5 |
| Asphaltenes | 10.6 | 13.9 | 7.2 | 15.3 | 12.9 | 15.5 | 16.1 |
| Preasphaltenes | 4.5 | 6.9 | 3.0 | 11.3 | 4.2 | 8.1 | 7.9 |
| $H_2$ CONSUMPTION, WT. % OF maf COAL | 8.8 | 8.1 | 9.1 | 7.3 | 8.4 | 7.9 | 7.8 |

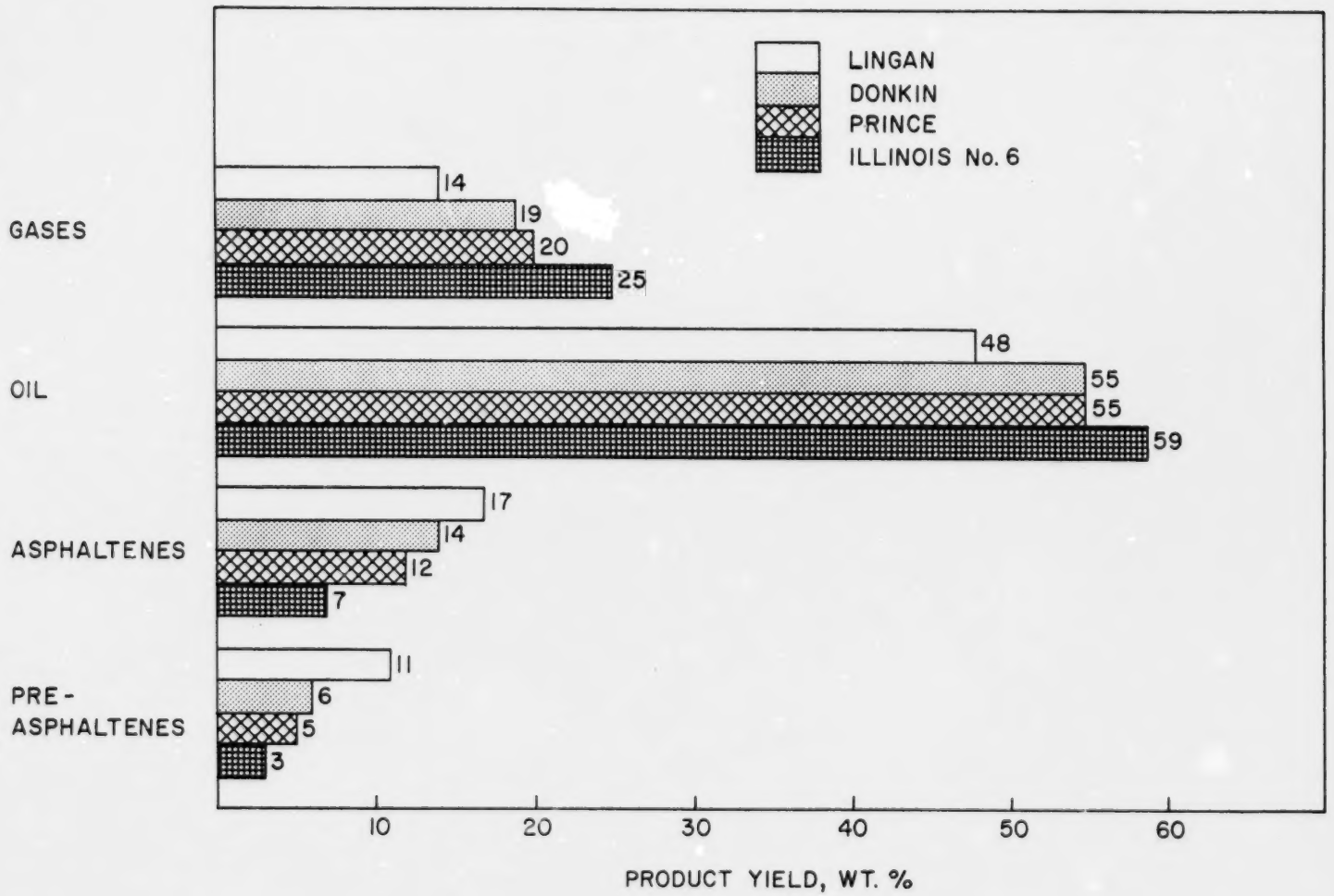
### 5.2.1 Coal Reactivity

The three Sydney area coals and Illinois No. 6 were tested at similar operating conditions in Run Nos. 8003, 8004, 8005 and 8011. The first and second-stage reactor temperatures were 400 and 425°C respectively. Slurry feedrate was targetted at 1.0 kg/hr and system pressure was maintained at 17.24 MPa. High coal conversion was achieved in all four experiments, 90 - 94%.

Product distribution varied considerably as a function of coal feedstock as illustrated in Figure 7. Gas yields ranged from 14% with Lingan Harbour seam coal to 25% with Illinois No. 6. Prince Hub and Donkin Harbour seam coals produced 55% oil compared with 48% from Lingan and 59% from Illinois No. 6. Lingan produced the highest yields of asphaltenes and preasphaltenes at 17 and 11% respectively. In fact, the three Sydney coals yielded higher levels of asphaltenic material than the American coal. Hydrogen consumption ranged from 6.6% in Run No. 8005 with Lingan coal to 9.1% in Run No. 8011 with Illinois No. 6.

The order of coal reactivity established is as follows: Illinois No. 6 > Prince Hub $\geq$ Donkin Harbour > Lingan Harbour. This order is quite similar to the trend in activity observed in earlier work under thermal/catalytic operating conditions. However, these experiments show product yields from the Prince Hub and Donkin Harbour coals to be essentially the same whereas the thermal/catalytic test work showed Prince coal to be slightly more reactive.

Figure 7 - PRODUCT DISTRIBUTION AS A FUNCTION OF COAL FEEDSTOCK IN CATALYTIC TWO-STAGE LIQUEFACTION



### 5.2.2 Effect of First-Stage Reactor Temperature

The first-stage reactor temperature was raised to 420°C in Run Nos. 8006, 8007 and 8008. The increase of 20°C in temperature resulted in slightly higher gas and oil yields from all three Nova Scotia coals. Hydrogen consumption increased as expected and the combined asphaltene/preasphaltene yield decreased moderately.

Figure 8 shows the effect of first-stage reactor temperature on conversion and raw product yields from Donkin Harbour seam coal. The data was obtained in Run Nos. 8012, 8015 and 8014 at 400, 420 and 440°C respectively. As the temperature was raised from 400 to 440°C; coal conversion remained fairly constant around 93%, gas make increased from 16.5 to 23.3%, oil yield rose from 49.8 to 53.2%, asphaltenes decreased from 15.3 to 12.9% and preasphaltenes decreased from 11.3 to 4.2%.

### 5.2.3 Influence of Total Pressure

Run Nos. 8010 and 8012 show the effect of operating pressure on process performance using Donkin coal. The former test was conducted at 20.68 MPa, the latter at 17.24 MPa. Coal conversion in both runs was 93%. Gas and oil yields were higher at 20.68 MPa, 20.4 and 52.1% respectively in Run No. 8010 compared with 16.5 and 49.8% in Run No. 8012. Preasphaltenes were substantially lower at 20.68 MPa, 6.9% compared with 11.3% at the lower pressure. Higher hydrogen consumption was measured in Run No. 8010 at 20.68 MPa.

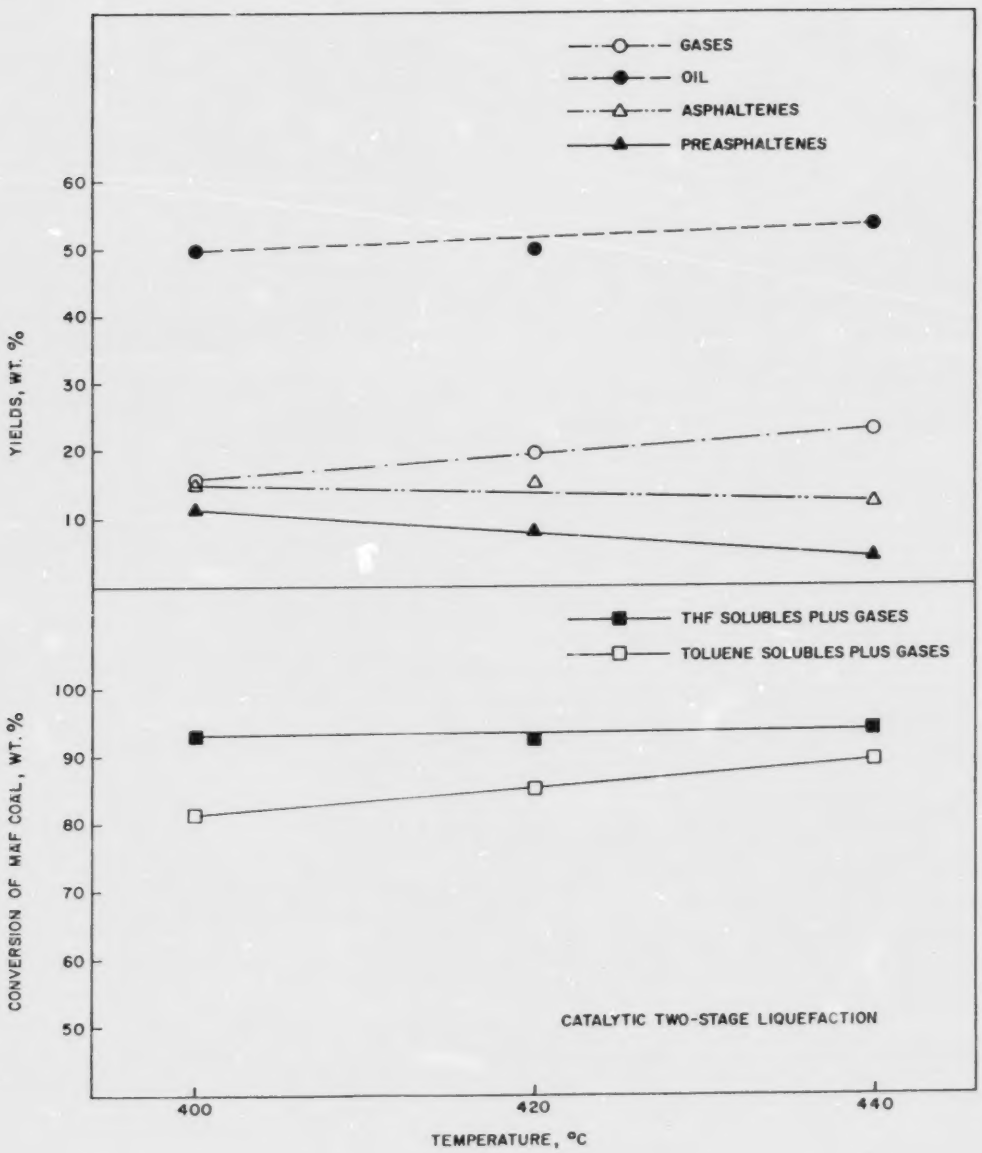



Figure 8 - EFFECT OF FIRST-STAGE CATALYTIC REACTOR TEMPERATURE ON COAL CONVERSION

### 5.2.4 Effect of Stirring Rate

In all of the experiments except Run No. 8009 the stirring rate was fixed at 800 RPM. In Run No. 8009 the stirring rate was increased to 1200 RPM. A comparison of results from Run Nos. 8008 and 8009 made with Prince coal shows that conversions and product distributions were quite similar. Oil yield was 56.9% at 800 RPM compared with 56.1% at 1200 RPM. Hydrogen consumption was measured at 8.8% in Run No. 8009 compared to 8.6% in Run No. 8008.

### 5.2.5 Influence of Catalyst Age

Data presented in Table 14 shows a decrease in catalyst activity with age. The four experiments were carried out with Donkin coal at two levels of temperature in the first-stage reactor, 400 and 420°C. A comparison of results from Run Nos. 8004 and 8012 at 400°C shows that process performance declined as catalyst in the first and second-stage reactors aged from 175 and 51 kg coal/kg catalyst respectively to 423 and 123 kg coal/kg catalyst. Oil yield decreased from 54.7% in the former run to 49.8% in the latter while preasphaltenes increased from 5.7 to 11.3% and hydrogen consumption declined from 7.6 to 7.3%. Similar trends were observed at a first-stage reactor temperature of 420°C in Run Nos. 8007 and 8015. Furthermore, as the catalyst aged the levels of sulphur and nitrogen in the pentane, toluene and THF soluble product fractions increased as shown in Table 15.

The occurrence of catalyst deactivation raises certain questions with regard to the reproducibility of experimental data and the validity of comparing experimental results. In response to these questions there are two relevant points. First, the experimental data is reproducible, as demonstrated by Run Nos. 8015 and 8016 presented in Table 13. Secondly, as the experimental program progressed the sequence of runs was controlled to minimize catalyst ageing between tests to be compared directly.

TABLE 14

RAW PRODUCT DISTRIBUTION AS A FUNCTION OF CATALYST AGE

| RUN NO.<br>COAL TESTED | 8004<br>DONKIN HR-8 | 8012<br>DONKIN HR-8 | 8007<br>DONKIN HR-8 | 8015<br>DONKIN HR-8 |
|---|---|---|---|---|
| Temperatures: °C | | | | |
| First-Stage Reactor | 400 | 400 | 420 | 420 |
| Second-Stage Reactor | 425 | 425 | 425 | 425 |
| Back Pressure, MPa | 17.24 | 17.24 | 17.24 | 17.24 |
| Slurry Feedrate, kg/hr | 1.041 | 1.049 | 1.019 | 0.999 |
| Hydrogen Treatrate, Wt. % Dry Coal | 19.3 | 19.1 | 19.7 | 20.1 |
| Catalyst Age, kg coal/kg catalyst | | | | |
| First-Stage Reactor | 175 | 423 | 268 | 486 |
| Second-Stage Reactor | 51 | 123 | 78 | 141 |
| CONVERSIONS, WT. % OF maf COAL | | | | |
| Pentane Solubles plus Gases | 73.5 | 66.3 | 77.3 | 69.4 |
| Toluene Solubles plus Gases | 87.2 | 81.6 | 89.4 | 84.9 |
| THF Solubles plus Gases | 92.9 | 92.9 | 93.7 | 93.0 |
| PRODUCT DISTRIBUTION, WT. % OF maf COAL | | | | |
| Gases | 18.8 | 16.5 | 19.6 | 19.9 |
| Oil | 54.7 | 49.8 | 57.7 | 49.5 |
| Asphaltenes | 13.7 | 15.3 | 12.1 | 15.5 |
| Preasphaltenes | 5.7 | 11.3 | 4.3 | 8.1 |
| $H_2$ CONSUMPTION, WT. % OF maf COAL | 7.6 | 7.3 | 8.7 | 7.9 |

TABLE 15

ELEMENTAL ANALYSIS ON PENTANE, TOLUENE AND THF SOLUBLE FRACTIONS FROM CATALYTIC TWO-STAGE LIQUEFACTION TESTS

| RUN NO. | 8002 | 8003 | 8004 | 8005 | 8006 | 8007 | 8008 |
|---|---|---|---|---|---|---|---|
| Pentane Soluble Fraction | | | | | | | |
| Carbon, Wt. % | 89.93 | 89.75 | 89.02 | 89.48 | 89.17 | 89.02 | 88.77 |
| Hydrogen, Wt. % | 9.11 | 9.34 | 9.08 | 9.08 | 9.24 | 9.18 | 9.39 |
| Nitrogen, Wt. % | 0.21 | 0.08 | 0.08 | 0.11 | 0.09 | 0.15 | 0.16 |
| Sulphur, Wt. % | 0.010 | 0.022 | 0.021 | 0.008 | 0.008 | 0.023 | 0.022 |
| Toluene Soluble Fraction | | | | | | | |
| Carbon, Wt. % | 88.80 | 88.89 | 89.43 | 89.24 | 89.56 | 89.59 | 89.27 |
| Hydrogen, Wt. % | 8.89 | 9.03 | 8.71 | 9.12 | 9.24 | 9.10 | 9.12 |
| Nitrogen, Wt. % | 0.30 | 0.13 | 0.11 | 0.19 | 0.26 | 0.21 | 0.19 |
| Sulphur, Wt. % | 0.020 | 0.025 | 0.037 | 0.015 | 0.012 | 0.032 | 0.025 |
| THF Soluble Fraction | | | | | | | |
| Carbon, Wt. % | 88.63 | 89.37 | 88.72 | 89.41 | 88.51 | 88.82 | 89.34 |
| Hydrogen, Wt. % | 8.99 | 8.98 | 8.73 | 9.12 | 9.05 | 9.10 | 9.08 |
| Nitrogen, Wt. % | 0.31 | 0.15 | 0.14 | 0.24 | 0.17 | 0.23 | 0.20 |
| Sulphur, Wt. % | 0.033 | 0.040 | 0.067 | 0.023 | 0.020 | 0.060 | 0.034 |

TABLE 15 (Continued)

ELEMENTAL ANALYSIS ON PENTANE, TOLUENE AND THF SOLUBLE FRACTIONS FROM CATALYTIC TWO-STAGE LIQUEFACTION TESTS

| RUN NO. | 8009 | 8010 | 8011 | 8012 | 8014 | 8015 | 8016 |
|---|---|---|---|---|---|---|---|
| Pentane Soluble Fraction | | | | | | | |
| Carbon, Wt. % | 88.44 | 89.58 | 89.50 | 89.37 | 88.45 | 89.50 | 89.02 |
| Hydrogen, Wt. % | 9.54 | 9.36 | 9.53 | 9.47 | 9.63 | 9.38 | 9.47 |
| Nitrogen, Wt. % | 0.14 | 0.13 | 0.15 | 0.17 | 0.15 | 0.16 | 0.15 |
| Sulphur, Wt. % | 0.023 | 0.026 | 0.025 | 0.027 | 0.026 | 0.025 | 0.026 |
| Toluene Soluble Fraction | | | | | | | |
| Carbon, Wt. % | 88.55 | 89.00 | 89.45 | 90.18 | 89.00 | 89.39 | 88.97 |
| Hydrogen, Wt. % | 9.22 | 9.36 | 9.32 | 9.32 | 9.46 | 9.11 | 9.35 |
| Nitrogen, Wt. % | 0.21 | 0.18 | 0.17 | 0.22 | 0.19 | 0.22 | 0.21 |
| Sulphur, Wt. % | 0.033 | 0.050 | 0.050 | 0.052 | 0.035 | 0.054 | 0.053 |
| THF Soluble Fraction | | | | | | | |
| Carbon, Wt. % | 89.19 | 88.57 | 88.38 | 89.64 | 88.58 | 88.77 | 88.57 |
| Hydrogen, Wt. % | 9.13 | 9.19 | 9.31 | 9.23 | 9.35 | 9.10 | 9.26 |
| Nitrogen, Wt. % | 0.18 | 0.22 | 0.20 | 0.28 | 0.23 | 0.26 | 0.27 |
| Sulphur, Wt. % | 0.041 | 0.072 | 0.067 | 0.092 | 0.083 | 0.092 | 0.083 |

## 5.3 Comparison of Catalytic Two-Stage and Two-Stage Thermal/Catalytic Process Performance

Although a limited quantity of data has been developed on catalytic two-stage liquefaction thusfar, it is interesting to make a comparison with two-stage thermal/catalytic liquefaction. Experimental results are presented in Table 16 for Prince Hub, Lingan Harbour and Donkin Harbour seam coals in both processing modes. The first-stage thermal dissolver was operated 20 - 30°C higher than the first-stage catalytic reactor in these tests. Total pressure was 17.24 MPa in the catalytic two-stage mode compared with 20.68 MPa in the thermal/catalytic mode.

Similar trends in product yields were observed with each of the coals. Catalytic two-stage liquefaction produced substantially less gas, moderately higher oil yields and significantly higher levels of preasphaltenes. Furthermore, hydrogen consumption was higher in the catalytic two-stage mode. For example, a comparison of Run No. 7014 (Donkin coal: thermal/catalytic) with Run No. 8007 (Donkin coal: catalytic/catalytic) showed the following shift in product distribution and hydrogen consumption:

- gases decreased from 27.7 to 19.6%
- oil increased from 53.5 to 57.7%
- asphaltenes increased from 11.4 to 12.1%
- preasphaltenes increased from 1.3 to 4.3%
- hydrogen consumption increased from 8.3 to 8.7%

Product yields are illustrated for the three Sydney area coals in Figure 9.

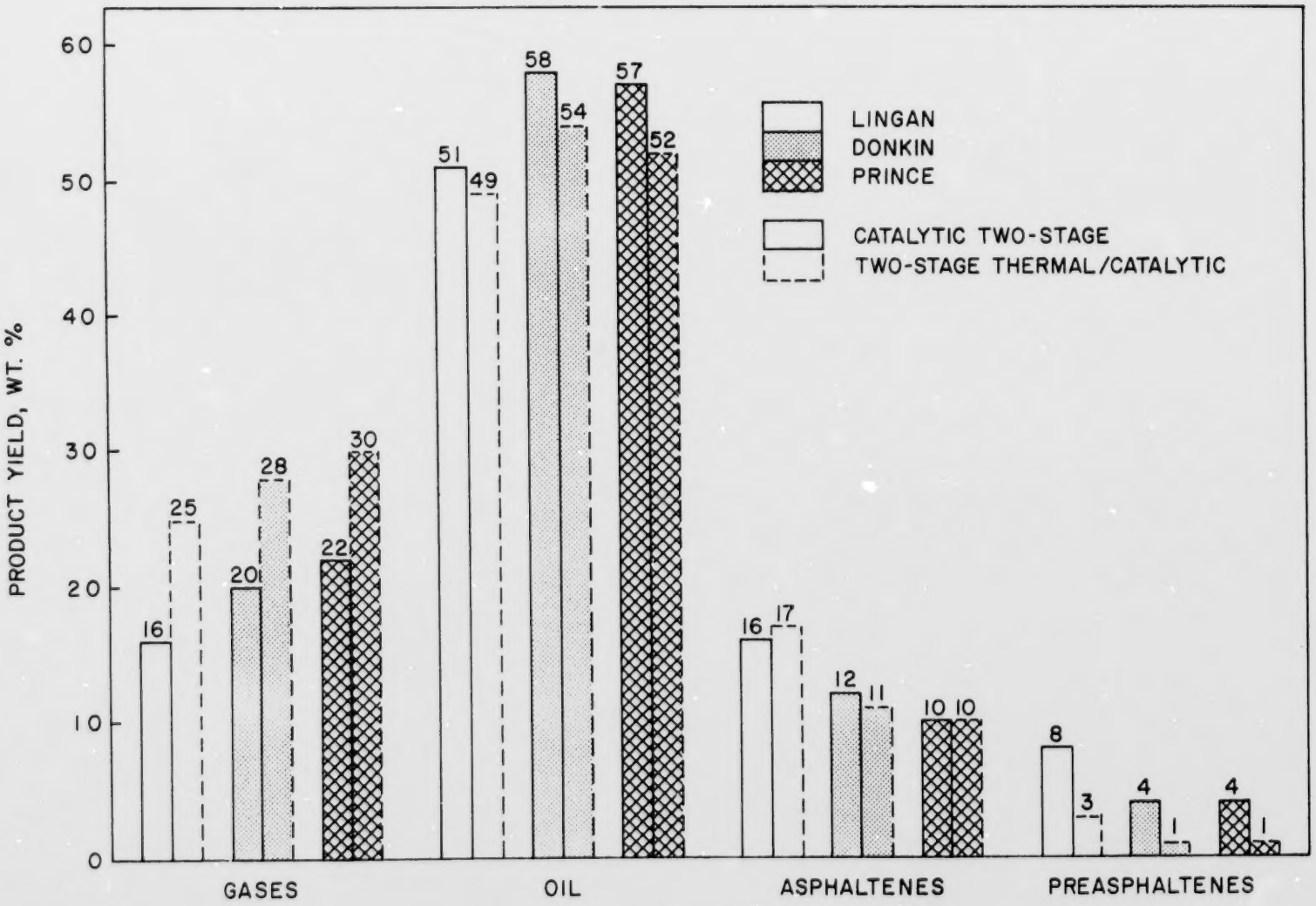
The higher oil yield and lower gas make associated with catalytic two-stage liquefaction is very promising. On the other hand, it seems that the thermal dissolver operating at higher temperature and pressure than the catalytic first-stage reactor yields less preasphaltenes. These large multi-ring compounds may be produced directly as the complex coal structure dissolves or indirectly via regressive reactions involving free-radicals. Regardless of the actual mechanism of formation, the presence of asphaltenic components will contribute to catalyst deactivation. Future work on catalytic two-stage processing will focus on the use of more effective catalyst and/or longer residence time in the first-stage reactor. The objective being to promote coal dissolution reactions and reduce the average size of coal-derived molecules.

TABLE 16

COMPARISON OF EXPERIMENTAL RESULTS FROM TWO-STAGE THERMAL/CATALYTIC AND CATALYTIC TWO-STAGE LIQUEFACTION

| RUN NO.<br>COAL TESTED | 8008<br>PRINCE HB-8 | 7016<br>PRINCE HB-7 | 8007<br>DONKIN HR-8 | 7014<br>DONKIN HR-7 | 8006<br>LINGAN HR-8 | 7015<br>LINGAN HR-7 |
|---|---|---|---|---|---|---|
| Temperatures: °C | | | | | | |
| First-Stage Reactor | 420 | 440 | 420 | 450 | 420 | 450 |
| Second-Stage Reactor | 425 | 425 | 425 | 425 | 425 | 425 |
| Back Pressure, MPa | 17.24 | 20.68 | 17.24 | 20.68 | 17.24 | 20.68 |
| $H_2$ Treatrate, Wt. % Dry Coal | 19.8 | 24.8 | 19.7 | 25.5 | 19.4 | 24.8 |
| Catalyst Age, kg coal/kg catalyst | | | | | | |
| First-Stage Reactor | 299 | - | 268 | - | 237 | - |
| Second-Stage Reactor | 87 | 94 | 78 | 76 | 69 | 85 |
| CONVERSIONS, WT. % OF maf COAL | | | | | | |
| Pentane Solubles plus Gases | 78.4 | 82.6 | 77.3 | 81.2 | 67.4 | 74.5 |
| Toluene Solubles plus Gases | 88.8 | 92.2 | 89.4 | 92.6 | 83.6 | 91.0 |
| THF Solubles plus Gases | 92.8 | 93.2 | 93.7 | 93.9 | 91.1 | 94.1 |
| PRODUCT DISTRIBUTION, WT. % OF maf COAL | | | | | | |
| Gases | 21.5 | 30.2 | 19.6 | 27.7 | 16.3 | 25.2 |
| Oil | 56.9 | 52.4 | 57.7 | 53.5 | 51.1 | 49.3 |
| Asphaltenes | 10.4 | 9.6 | 12.1 | 11.4 | 16.2 | 16.5 |
| Preasphaltenes | 4.0 | 1.0 | 4.3 | 1.3 | 7.5 | 3.1 |
| $H_2$ CONSUMPTION, WT. % OF maf COAL | 8.6 | 7.1 | 8.7 | 8.3 | 7.5 | 6.6 |

Figure 9 - RAW PRODUCT YIELDS AS A FUNCTION OF COAL FEEDSTOCK AND LIQUEFACTION PROCESSING MODE



## CONCLUSIONS AND RECOMMENDATIONS

A research program on catalytic two-stage liquefaction of Nova Scotia bituminous coals has been initiated. Bench-scale studies were conducted to assess the effects of operating conditions on product yields from three Sydney area coals. Experiments were conducted in a once-through mode using hydrogenated coal-derived oil as reaction solvent. In addition, the planned extension of a two-stage thermal/catalytic liquefaction run with solvent recycle was completed.

During the previous phase of work a two-stage thermal/catalytic recycle test was run successfully for 6 days using Prince Hub seam coal. The extension of this run was halted after 4½ days due to unfavourable experimental results. As the run progressed hydrogen consumption decreased and the specific gravity of the recycle liquid steadily increased. Coal conversion over the last 12-hour operating period averaged 85%. The net $C_4^+$ liquid yield was 57%. Yields of naphtha and light gas oil were 36 and 38 wt.% respectively.

Analysis of product and recycle streams indicate that as the run progressed the donor capacity of the recycle liquid decreased. As solvent quality decreased; coal conversion decreased, liquid product yield fell off and product quality declined. Furthermore, as the concentration of heavy asphaltenic components in the recycle liquid increased so did its propensity to coke. This resulted in extensive coke formation in the second-stage catalytic reactor which was operating at high temperature and low slurry space velocity.

A total of fourteen catalytic two-stage experiments were conducted with Amocat 1C nickel-molybdenum catalyst. The runs were made to investigate coal reactivity and the effects of first-stage reactor temperature, pressure, stirring rate and catalyst age on raw product yields. Four coals were tested: Lingan Harbour, Donkin Harbour, Prince Hub and Illinois No. 6.

High coal conversions, 90 - 94%, were achieved with all four coals at a first-stage reactor temperature of 400°C. However, product distribution varied widely as a function of coal feedstock. Gas yield ranged from 14% with Lingan Harbour seam coal to 25% with Illinois No. 6. Prince Hub and Donkin Harbour seam coals produced 55% oil compared with 48% from Lingan and 59% from Illinois No. 6. Lingan produced the highest yields of asphaltenes and preasphaltenes at 17 and 11% respectively. The three Sydney area coals produced substantially higher levels of asphaltenes and preasphaltenes than the American coal. Hydrogen consumption ranged from 6.6% with Lingan coal to 9.1% with Illinois No. 6.

An increase in first-stage temperature of 20° to 420°C produced slightly higher gas and oil yields from all three Nova Scotia coals. Hydrogen consumption increased as expected and combined asphaltene/preasphaltene yields decreased moderately. The order of coal reactivity established is as follows: Illinois No. 6 > Prince Hub $\geq$ Donkin Harbour > Lingan Harbour.

Experiments with Donkin coal showed that as first-stage temperature was raised from 400 to 440°C; coal conversion was firm at 93%, gas make increased from 17 to 23%, oil yield rose from 50 to 53%, asphaltenes decreased from 15 to 13% and preasphaltenes decreased from 11 to 4%. Increasing the operating pressure from 17.24 to 20.68 MPa at 400°C resulted in slightly higher gas and oil yields while the preasphaltene fraction was reduced from 11 to 7%. Tests with Prince coal at stirring rates of 800 and 1200 RPM produced similar coal conversion and raw product yields.

Catalyst activity was found to decrease rapidly as a function of age. In tests with Donkin coal at 400°C, oil yield decreased from 55 to 50% as the first and second-stage catalyst aged from 175 and 51 kg coal/kg catalyst respectively to 423 and 123 kg coal/kg catalyst. Furthermore, preasphaltenes increased from 6 to 11% and hydrogen consumption decreased from 7.6 to 7.3%.

A comparison of experimental results showed catalytic two-stage liquefaction to produce substantially lower gas yields and moderately higher oil yields than two-stage thermal/catalytic liquefaction. However, it seems that the thermal dissolver operating at higher temperature and pressure than the catalytic first-stage reactor yields less preasphaltenes. These large multi-ring compounds may be produced directly as the complex coal structure dissolves or indirectly via regressive reactions involving free-radicals. Regardless of the actual mechanism of formation, the presence of heavy asphaltenic components will contribute to catalyst deactivation.

Based on the results from this work the following recommendations for future work are proposed:

(a) Continue the bench program on catalytic two-stage liquefaction. Conduct short-term exploratory trials to investigate the use of additional catalysts and/or longer slurry residence time in the first-stage reactor to reduce asphaltene production.

(b) Undertake catalytic two-stage liquefaction tests with liquid recycle to determine ultimate yields, product composition and catalyst ageing characteristics.

(c) Revisit two-stage thermal/catalytic liquefaction to assess the potential of process modifications designed to control asphaltenes content in the recycle stream.

(d) Conduct catalyst regeneration studies to determine the extent to which used catalyst can be reactivated for recycle.

At present, conventional coal liquefaction is not economically viable in Eastern Canada. Technological advances in the areas of process and/or plant hardware are required to improve the economics of converting coal into premium quality liquid fuels. Two-stage liquefaction offers the potential to enhance distillate yields and improve hydrogen utilization. However, extensive test work is required

on two-stage process configurations applied to Sydney area coals to determine the extent to which these improvements can be realized.

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# APPENDIX A

## CONVERSION CALCULATIONS

APPENDIX A

## CONVERSION CALCULATIONS FOR CONTINUOUS COAL LIQUEFACTION EXPERIMENTS

Calculations are based on operations during the steady state sampling period.

### CONVERSIONS. WT.%:

$$\text{Pentane sol. + gases} = \frac{(\text{maf coal charged}) - (\text{organic pentane insol.})}{\text{maf coal charged}} \times 100$$

$$\text{Toluene sol. + gases} = \frac{(\text{maf coal charged}) - (\text{organic toluene insol.})}{\text{maf coal charged}} \times 100$$

$$\text{THF sol. + gases} = \frac{(\text{maf coal charged}) - (\text{organic THF insol.})}{\text{maf coal charged}} \times 100$$

$$\text{Gases} = \frac{\text{product gases}}{\text{maf coal charged}} \times 100$$

### DEFINITION OF PRODUCTS

Oil = pentane solubles

Asphaltenes = toluene solubles - pentane solubles

Preasphaltenes = THF solubles - toluene solubles

# APPENDIX B

## SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE FOR TWO-STAGE THERMAL/CATALYTIC LIQUEFACTION EXPERIMENTS

## SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE FOR RCL-8000

| | | | |
|---|---|---|---|
| OPERATING PERIOD, HRS. | 0 - 12 | 12 - 24 | 24 - 36 |
| COAL TESTED | PRINCE HB-8/HB-7 | PRINCE HB-8/HB-7 | PRINCE HB-8 |
| CATALYST TESTED | AMOCAT 1C | AMOCAT 1C | AMOCAT 1C |
| A. OPERATING CONDITIONS | | | |
| Unit back pressure, MPa | 20.68 | 20.68 | 20.68 |
| Temperatures, °C: | | | |
| Thermal Dissolver | 440 | 440 | 440 |
| Catalytic Reactor | 435 | 435 | 435 |
| Slurry Preheater | 250 | 250 | 250 |
| Hot Separator | 300 | 300 | 300 |
| Feedrates: | | | |
| Slurry, kg/hr | 1.010 | 0.980 | 0.997 |
| Raw Coal, kg/hr | 0.283 | 0.274 | 0.279 |
| Hydrogen, L/hr | 614. | 614. | 614. |
| SHSV, $hr^{-1}$; Thermal Dissolver | 1.00 | 1.00 | 1.00 |
| Catalytic Reactor | 0.33 | | |
| Hydrogen Treatrate, Wt. % Dry Coal | 20.2 | 20.8 | 20.5 |
| Catalyst Age (kg coal/kg catalyst) | 261 | 271 | 281 |
| B. CONVERSIONS OF maf COAL, WT. % | | | |
| THF Solubles plus Gases | 92.9 | 91.6 | 92.6 |
| C. PRODUCT YIELDS ON maf COAL, WT. % | | | |
| $CH_4$ | 5.4 | 5.3 | 5.3 |
| $C_2H_6$ | 5.9 | 5.1 | 5.2 |
| $C_3H_8$ | 4.0 | 3.7 | 3.6 |
| $C_4H_{10}$ | 3.7 | 3.7 | 3.4 |
| CO | 0.6 | 0.5 | 0.7 |
| $H_2S$ | 1.9 | 1.7 | 1.5 |
| $H_2O$ | 8.4 | 8.7 | 7.6 |
| $NH_3$ | 0.8 | 0.8 | 0.7 |
| Oil + Asphaltenes + Preasphaltenes | 62.2 | 62.1 | 64.6 |
| Unreacted Coal | 7.1 | 8.4 | 7.4 |
| D. $H_2$ CONSUMPTION, WT. % OF maf COAL | 7.2 | 7.4 | 6.5 |

## SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE FOR RCL-8000

| | | | |
|---|---|---|---|
| OPERATING PERIOD, HRS. | 36 - 48 | 48 - 60 | 60 - 72 |
| COAL TESTED | PRINCE HB-8 | PRINCE HB-8 | PRINCE HB-8 |
| CATALYST TESTED | AMOCAT 1C | AMOCAT 1C | AMOCAT 1C |
| A. OPERATING CONDITIONS | | | |
| Unit back pressure, MPa | 20.68 | 20.68 | 20.68 |
| Temperatures, °C: | | | |
| Thermal Dissolver | 440 | 450 | 455 |
| Catalytic Reactor | 435 | 435 | 440/445 |
| Slurry Preheater | 250 | 250 | 250 |
| Hot Separator | 300 | 300 | 300 |
| Feedrates: | | | |
| Slurry, kg/hr | 0.996 | 0.998 | 0.994 |
| Raw Coal, kg/hr | 0.279 | 0.279 | 0.278 |
| Hydrogen, L/hr | 614. | 614. | 614. |
| SHSV, $hr^{-1}$; Thermal Dissolver | 1.00 | 1.00 | 1.00 |
| Catalytic Reactor | | | |
| Hydrogen Treatrate, Wt. % Dry Coal | 20.5 | 20.5 | 20.5 |
| Catalyst Age (kg coal/kg catalyst) | 291 | 301 | 311 |
| B. CONVERSIONS OF maf COAL, WT. % | | | |
| THF Solubles plus Gases | 91.3 | 91.5 | 90.8 |
| C. PRODUCT YIELDS ON maf COAL, WT. % | | | |
| $CH_4$ | 5.5 | 6.9 | 9.4 |
| $C_2H_6$ | 5.5 | 5.5 | 7.2 |
| $C_3H_8$ | 4.0 | 4.7 | 5.7 |
| $C_4H_{10}$ | 3.5 | 3.7 | 4.3 |
| CO | 0.7 | 0.6 | 0.9 |
| $H_2S$ | 1.8 | 1.5 | 1.8 |
| $H_2O$ | 6.8 | 7.0 | 7.2 |
| $NH_3$ | 0.6 | 0.6 | 0.6 |
| Oil + Asphaltenes + Preasphaltenes | 62.9 | 61.0 | 53.7 |
| Unreacted Coal | 8.7 | 8.5 | 9.2 |
| D. $H_2$ CONSUMPTION, WT. % OF maf COAL | 6.1 | 6.1 | 6.6 |

## SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE FOR RCL-8000

| | | | |
|---|---|---|---|
| OPERATING PERIOD, HRS. | 72 - 84 | 84 - 96 | 96 - 108 |
| COAL TESTED | PRINCE HB-8 | PRINCE HB-8 | PRINCE HB-8 |
| CATALYST TESTED | AMOCAT 1C | AMOCAT 1C | AMOCAT 1C |
| A. OPERATING CONDITIONS | | | |
| Unit back pressure, MPa | 20.68 | 20.68 | 20.68 |
| Temperatures, °C: | | | |
| Thermal Dissolver | 455 | 450 | 450 |
| Catalytic Reactor | 445/440 | 435 | 435 |
| Slurry Preheater | 250 | 250 | 250 |
| Hot Separator | 300 | 300 | 300 |
| Feedrates: | | | |
| Slurry, kg/hr | 0.900 | 1.003 | 0.993 |
| Raw Coal, kg/hr | 0.252 | 0.281 | 0.278 |
| Hydrogen, L/hr | 614. | 614. | 614. |
| SHSV, $hr^{-1}$; Thermal Dissolver | 1.0 | 1.00 | 1.00 |
| Catalytic Reactor | | | |
| Hydrogen Treatrate, Wt. % Dry Coal | 20.3 | 20.3 | 20.5 |
| Catalyst Age (kg coal/kg catalyst) | 321 | 331 | 341 |
| B. CONVERSIONS OF maf COAL, WT. % | | | |
| THF Solubles plus Gases | 89.9 | 86.3 | 85.0 |
| C. PRODUCT YIELDS ON maf COAL, WT. % | | | |
| $CH_4$ | 10.0 | 7.8 | 7.6 |
| $C_2H_6$ | 7.7 | 5.4 | 5.1 |
| $C_3H_8$ | 6.0 | 4.3 | 4.4 |
| $C_4H_{10}$ | 4.1 | 3.5 | 3.3 |
| CO | 0.9 | 0.8 | 1.0 |
| $H_2S$ | 1.8 | 1.7 | 1.6 |
| $H_2O$ | 6.8 | 6.6 | 6.7 |
| $NH_3$ | 0.6 | 0.5 | 0.4 |
| Oil + Asphaltenes + Preasphaltenes | 52.0 | 55.7 | 54.9 |
| Unreacted Coal | 10.1 | 13.7 | 15.0 |
| D. $H_2$ CONSUMPTION, WT. % OF maf COAL | 6.1 | 5.7 | 5.8 |

SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE
FOR TWO-STAGE THERMAL/CATALYTIC LIQUEFACTION TESTS

| RUN NO. | 8001-8-10 | 8001-10-14 | 8001-14-16 |
|---|---|---|---|
| COAL TESTED | LINGAN HR-7 | LINGAN HR-7 | LINGAN HR-7 |
| CATALYST TESTED | AMOCAT 1C | AMOCAT 1C | AMOCAT 1C |
| **A. OPERATING CONDITIONS** | | | |
| Unit back pressure, MPa | 20.68 | 20.68 | 20.68 |
| Temperatures, °C: | | | |
| Thermal Dissolver | 460 | 460 | 460 |
| Catalytic Reactor | 435 | 435 | 435 |
| Slurry Preheater | 250 | 250 | 250 |
| Hot Separator | 300 | 300 | 300 |
| Feedrates: | | | |
| Slurry, kg/hr | 1.026 | 1.011 | 1.003 |
| Raw Coal, kg/hr | 0.277 | 0.273 | 0.271 |
| Hydrogen, L/hr | 740. | 740. | 740. |
| SHSV, $hr^{-1}$; Thermal Dissolver | 1.00 | 1.00 | 1.00 |
| Catalytic Reactor | | | |
| Hydrogen Treatrate, Wt. % Dry Coal | 24.4 | 24.7 | 24.9 |
| Catalyst Age (kg coal/kg catalyst) | 343 | 346 | 348 |
| **B. CONVERSIONS OF maf COAL, WT. %** | | | |
| Pentane Solubles plus Gases | 59.1 | 59.5 | 57.6 |
| Toluene Solubles plus Gases | 82.6 | 80.9 | 81.3 |
| THF Solubles plus Gases | 91.6 | 91.3 | 91.4 |
| **C. PRODUCT YIELDS ON maf COAL, WT. %** | | | |
| $CH_4$ | 7.7 | 7.9 | 7.6 |
| $C_2H_6$ | 7.9 | 7.5 | 7.1 |
| $C_3H_8$ | 5.5 | 5.4 | 5.9 |
| $C_4H_{10}$ | 4.3 | 4.1 | 4.8 |
| CO | 0.9 | 1.3 | 1.0 |
| $H_2S$ | 1.2 | 1.1 | 0.9 |
| $H_2O$ | 4.7 | 4.5 | 4.1 |
| $NH_3$ | 0.4 | 0.4 | 0.4 |
| Oil | 26.5 | 27.3 | 25.8 |
| Asphaltenes | 23.5 | 21.4 | 23.7 |
| Preasphaltenes | 9.0 | 10.4 | 10.1 |
| Unreacted Coal | 8.4 | 8.7 | 8.6 |
| **D. $H_2$ CONSUMPTION, WT. % OF maf COAL** | 5.9 | 5.9 | 5.5 |

# APPENDIX C

## MATERIAL BALANCES FOR TWO-STAGE THERMAL/CATALYTIC LIQUEFACTION EXPERIMENTS

## MATERIAL BALANCES FOR RCL-8000

| OPERATING PERIOD, HRS. | 0 - 12 | 12 - 24 | 24 - 36 |
|---|---|---|---|
| COAL TESTED | PRINCE HB-8/HB-7 | PRINCE HB-8/HB-7 | PRINCE HB-8 |
| CATALYST TESTED | AMOCAT 1C | AMOCAT 1C | AMOCAT 1C |
| MOISTURE, WT. % | 3.26 | 3.24 | 3.22 |
| ASH, WT. % DRY BASIS | 7.48 | 6.67 | 5.86 |
| INPUT (g): | | | |
| $H_2O$ | 110.8 | 106.6 | 108.1 |
| Ash | 245.9 | 212.4 | 190.3 |
| maf Coal | 3041. | 2972. | 3058. |
| Solvent | 8738. | 8463. | 8631. |
| $H_2$ | 664.7 | 663.6 | 664.7 |
| Total | 12800. | 12418. | 12652. |
| OUTPUT (g): | | | |
| $H_2O$ | 366.8 | 365.3 | 340.9 |
| Ash | 140.5 | 173.1 | 124.6 |
| Organic THF Insolubles[1] | 215.3 | 249.1 | 226.5 |
| THF Solubles[2] | 11020. | 10398. | 10631. |
| $H_2$ | 445.5 | 445.1 | 466.2 |
| Other Gases | 678.4 | 617.4 | 624.3 |
| Total | 12867. | 12248. | 12414. |
| TOTAL MATERIAL RECOVERY, WT. % | 100.5 | 98.6 | 98.1 |

1 - Unreacted Coal
2 - Oil, Asphaltenes and Preasphaltenes

## MATERIAL BALANCES FOR RCL-8000

| OPERATING PERIOD, HRS. | 36 - 48 | 48 - 60 | 60 - 72 |
|---|---|---|---|
| COAL TESTED | PRINCE HB-8 | PRINCE HB-8 | PRINCE HB-8 |
| CATALYST TESTED | AMOCAT 1C | AMOCAT 1C | AMOCAT 1C |
| MOISTURE, WT. % | 3.22 | 3.22 | 3.22 |
| ASH, WT. % DRY BASIS | 5.86 | 5.86 | 5.86 |
| INPUT (g): | | | |
| $H_2O$ | 107.8 | 107.9 | 107.5 |
| Ash | 189.8 | 190.1 | 189.3 |
| maf Coal | 3049. | 3054. | 3041. |
| Solvent | 8605. | 8620. | 8584. |
| $H_2$ | 663.6 | 663.6 | 663.6 |
| Total | 12615. | 12636. | 12585. |
| OUTPUT (g): | | | |
| $H_2O$ | 315.4 | 320.2 | 327.2 |
| Ash | 138.1 | 126.3 | 100.0 |
| Organic THF Insolubles[1] | 264.0 | 258.5 | 279.5 |
| THF Solubles[2] | 10389. | 10360. | 10029. |
| $H_2$ | 476.6 | 477.1 | 462.6 |
| Other Gases | 661.0 | 716.6 | 912.6 |
| Total | 12244. | 12259. | 12111. |
| TOTAL MATERIAL RECOVERY, WT. % | 97.1 | 97.0 | 96.2 |

1 - Unreacted Coal
2 - Oil, Asphaltenes and Preasphaltenes

## MATERIAL BALANCES FOR RCL-8000

| OPERATING PERIOD, HRS. | 72 - 84 | 84 - 96 | 96 - 108 |
|---|---|---|---|
| COAL TESTED | PRINCE HB-8 | PRINCE HB-8 | PRINCE HB-8 |
| CATALYST TESTED | AMOCAT 1C | AMOCAT 1C | AMOCAT 1C |
| MOISTURE, WT. % | 3.22 | 3.22 | 3.22 |
| ASH, WT. % DRY BASIS | 5.86 | 5.86 | 5.86 |
| INPUT (g): | | | |
| $H_2O$ | 108.8 | 108.6 | 107.5 |
| Ash | 191.7 | 191.2 | 189.3 |
| maf Coal | 3080. | 3072. | 3041. |
| Solvent | 8692. | 8670. | 8583. |
| $H_2$ | 665.2 | 663.6 | 663.6 |
| Total | 12738. | 12705. | 12584. |
| OUTPUT (g): | | | |
| $H_2O$ | 319.5 | 311.1 | 311.9 |
| Ash | 99.3 | 134.0 | 154.0 |
| Organic THF Insolubles[1] | 310.9 | 421.2 | 454.8 |
| THF Solubles[2] | 10101. | 10173. | 10668. |
| $H_2$ | 476.2 | 489.2 | 486.7 |
| Other Gases | 958.4 | 736.2 | 710.2 |
| Total | 12265. | 12265. | 12786. |
| TOTAL MATERIAL RECOVERY, WT. % | 96.3 | 96.5 | 101.6 |

1 - Unreacted Coal
2 - Oil, Asphaltenes and Preasphaltenes

MATERIAL BALANCES FOR TWO STAGE
THERMAL/CATALYTIC LIQUEFACTION TESTS

| RUN NO. | 8001-8-10 | 8001-10-14 | 8001-14-16 |
|---|---|---|---|
| COAL TESTED | LINGAN HR-7 | LINGAN HR-7 | LINGAN HR-7 |
| CATALYST TESTED | AMOCAT 1C | AMOCAT 1C | AMOCAT 1C |
| MOISTURE, WT. % | 1.28 | 1.28 | 1.28 |
| ASH, WT. % DRY BASIS | 3.42 | 3.42 | 3.42 |
| HOURS ON STREAM | 16.02 | 16.02 | 16.02 |
| DURATION OF BALANCE PERIOD, HRS. | 2.03 | 2.02 | 2.06 |
| INPUT (g): | | | |
| $H_2O$ | 7.0 | 14.1 | 7.0 |
| Ash | 18.5 | 37.1 | 18.5 |
| maf Coal | 523. | 1048. | 523. |
| Solvent | 1483. | 2972. | 1483. |
| $H_2$ | 131.9 | 268.2 | 134.9 |
| Total | 2163. | 4339. | 2166. |
| OUTPUT (g): | | | |
| $H_2O$ | 31.5 | 61.3 | 28.4 |
| Ash | 16.9 | 34.2 | 18.9 |
| Organic THF Insolubles[1] | 43.8 | 91.7 | 44.7 |
| THF Solubles[2] | 1789. | 3569. | 1837. |
| $H_2$ | 101.1 | 206.8 | 106.0 |
| Other Gases | 146.2 | 289.3 | 144.8 |
| Total | 2129. | 4252. | 2180. |
| TOTAL MATERIAL RECOVERY, WT. % | 98.4 | 98.0 | 100.6 |

1 - Unreacted Coal
2 - Oil, Asphaltenes and Preasphaltenes

# APPENDIX D

## SUMMARY OF OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC TWO-STAGE LIQUEFACTION EXPERIMENTS

OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

| RUN NO.<br>COAL TESTED<br>CATALYST TESTED | 8002<br>LINGAN HR-6<br>AMOCAT 1C | 8003<br>PRINCE HB-8<br>AMOCAT 1C | 8004<br>DONKIN HR-8<br>AMOCAT 1C | 8005<br>LINGAN HR-8<br>AMOCAT 1C |
|---|---|---|---|---|
| A. OPERATING CONDITIONS | | | | |
| Unit back pressure, MPa | 17.24 | 17.24 | 17.24 | 17.24 |
| Temperatures, °C: | | | | |
| First-Stage Reactor | 415 | 400 | 400 | 400 |
| Second-Stage Reactor | 425 | 425 | 425 | 425 |
| Hot Separator | 300 | 300 | 300 | 300 |
| Feedrates: | | | | |
| Slurry, kg/hr | 1.546 | 1.006 | 1.041 | 1.048 |
| Raw Coal, kg/hr | 0.433 | 0.282 | 0.291 | 0.293 |
| Hydrogen, L/hr | 462. | 600. | 612. | 618. |
| SHSV, $hr^{-1}$; First-Stage | 1.50 | 1.00 | 1.00 | 1.00 |
| Second-Stage | 0.50 | 0.33 | 0.33 | 0.33 |
| Hydrogen Treatrate, Wt. % Dry Coal | 9.7 | 19.8 | 19.3 | 19.1 |
| Catalyst Age, (kg coal/kg catalyst): | | | | |
| First-Stage Reactor | 107 | 144 | 175 | 206 |
| Second-Stage Reactor | 31 | 42 | 51 | 60 |
| B. CONVERSIONS OF maf COAL, WT. % | | | | |
| Pentane Solubles plus Gases | 57.1 | 75.0 | 73.5 | 61.8 |
| Toluene Solubles plus Gases | 77.8 | 87.3 | 87.2 | 78.9 |
| THF Solubles plus Gases | 90.5 | 92.2 | 92.9 | 90.0 |
| C. PRODUCT YIELDS ON maf COAL, WT. % | | | | |
| $CH_4$ | 2.0 | 2.3 | 2.3 | 2.2 |
| $C_2H_6$ | 1.8 | 2.2 | 2.5 | 2.4 |
| $C_3H_8$ | 1.2 | 1.7 | 1.6 | 1.3 |
| $C_4H_{10}$ | 1.5 | 2.3 | 2.5 | 2.1 |
| CO | 0.1 | 0.2 | 0.1 | 0.1 |
| $H_2S$ | 0.5 | 1.3 | 1.5 | 0.5 |
| $H_2O$ | 5.2 | 8.5 | 7.3 | 4.6 |
| $NH_3$ | 0.7 | 1.1 | 1.0 | 0.9 |
| Oil | 44.1 | 55.4 | 54.7 | 47.7 |
| Asphaltenes | 20.7 | 12.3 | 13.7 | 17.1 |
| Preasphaltenes | 12.7 | 4.9 | 5.7 | 11.1 |
| Unreacted Coal | 9.5 | 7.8 | 7.1 | 10.0 |
| D. $H_2$ CONSUMPTION, WT. % maf COAL | 5.2 | 8.3 | 7.6 | 6.6 |

## OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

| RUN NO.<br>COAL TESTED<br>CATALYST TESTED | 8006<br>LINGAN HR-8<br>AMOCAT 1C | 8007<br>DONKIN HR-8<br>AMOCAT 1C | 8008<br>PRINCE HB-8<br>AMOCAT 1C | 8009<br>PRINCE HB-8<br>AMOCAT 1C |
|---|---|---|---|---|
| **A. OPERATING CONDITIONS** | | | | |
| Unit back pressure, MPa | 17.24 | 17.24 | 17.24 | 17.24 |
| Temperatures, °C: | | | | |
| First-Stage Reactor | 420 | 420 | 420 | 420 |
| Second-Stage Reactor | 425 | 425 | 425 | 425 |
| Hot Separator | 300 | 300 | 300 | 300 |
| Feedrates: | | | | |
| Slurry, kg/hr | 1.037 | 1.019 | 1.028 | 1.000 |
| Raw Coal, kg/hr | 0.290 | 0.285 | 0.288 | 0.280 |
| Hydrogen, L/hr | 618. | 612. | 612. | 612. |
| SHSV, $hr^{-1}$; First-Stage | 1.00 | 1.00 | 1.00 | 1.00 |
| Second-Stage | 0.33 | 0.33 | 0.33 | 0.33 |
| Hydrogen Treatrate, Wt. % Dry Coal | 19.4 | 19.7 | 19.8 | 20.3 |
| Catalyst Age, (kg coal/kg catalyst): | | | | |
| First-Stage Reactor | 237 | 268 | 299 | 330 |
| Second-Stage Reactor | 69 | 78 | 87 | 96 |
| **B. CONVERSIONS OF maf COAL, WT. %** | | | | |
| Pentane Solubles plus Gases | 67.4 | 77.3 | 78.4 | 77.2 |
| Toluene Solubles plus Gases | 83.6 | 89.4 | 88.8 | 87.8 |
| THF Solubles plus Gases | 91.1 | 93.7 | 92.8 | 92.3 |
| **C. PRODUCT YIELDS ON maf COAL, WT. %** | | | | |
| $CH_4$ | 3.0 | 2.5 | 2.7 | 2.5 |
| $C_2H_6$ | 2.9 | 3.0 | 2.7 | 2.6 |
| $C_3H_8$ | 1.8 | 2.0 | 2.1 | 1.4 |
| $C_4H_{10}$ | 2.4 | 2.5 | 2.5 | 2.1 |
| CO | 0.4 | 0.1 | 0.3 | 0.2 |
| $H_2S$ | 0.5 | 1.2 | 1.4 | 1.5 |
| $H_2O$ | 4.4 | 7.2 | 8.7 | 9.6 |
| $NH_3$ | 0.9 | 1.1 | 1.1 | 1.2 |
| Oil | 51.1 | 57.7 | 56.9 | 56.1 |
| Asphaltenes | 16.2 | 12.1 | 10.4 | 10.6 |
| Preasphaltenes | 7.5 | 4.3 | 4.0 | 4.5 |
| Unreacted Coal | 8.9 | 6.3 | 7.2 | 7.7 |
| **D. $H_2$ CONSUMPTION, WT. % maf COAL** | 7.5 | 8.7 | 8.6 | 8.8 |

## OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

| RUN NO. | 8010 | 8011 | 8012 | 8014 |
|---|---|---|---|---|
| COAL TESTED | DONKIN HR-8 | ILLINOIS NO. 6 | DONKIN HR-8 | DONKIN HR-8 |
| CATALYST TESTED | AMOCAT 1C | AMOCAT 1C | AMOCAT 1C | AMOCAT 1C |
| **A. OPERATING CONDITIONS** | | | | |
| Unit back pressure, MPa | 20.68 | 17.24 | 17.24 | 17.24 |
| Temperatures, °C: | | | | |
| First-Stage Reactor | 400 | 400 | 400 | 440 |
| Second-Stage Reactor | 425 | 425 | 425 | 425 |
| Hot Separator | 300 | 300 | 300 | 300 |
| Feedrates: | | | | |
| Slurry, kg/hr | 1.028 | 1.040 | 1.049 | 1.032 |
| Raw Coal, kg/hr | 0.288 | 0.291 | 0.294 | 0.289 |
| Hydrogen, L/hr | 612. | 606. | 612. | 618. |
| SHSV, $hr^{-1}$; First-Stage | 1.00 | 1.00 | 1.00 | 1.00 |
| Second-Stage | 0.33 | 0.33 | 0.33 | 0.33 |
| Hydrogen Treatrate, Wt. % Dry Coal | 19.5 | 19.3 | 19.1 | 19.6 |
| Catalyst Age, (kg coal/kg catalyst): | | | | |
| First-Stage Reactor | 361 | 392 | 423 | 454 |
| Second-Stage Reactor | 105 | 114 | 123 | 132 |
| **B. CONVERSIONS OF maf COAL, WT. %** | | | | |
| Pentane Solubles plus Gases | 72.5 | 84.1 | 66.3 | 76.5 |
| Toluene Solubles plus Gases | 86.4 | 91.3 | 81.6 | 89.4 |
| THF Solubles plus Gases | 93.3 | 94.3 | 92.9 | 93.6 |
| **C. PRODUCT YIELDS ON maf COAL, WT. %** | | | | |
| $CH_4$ | 2.2 | 2.0 | 2.1 | 3.4 |
| $C_2H_6$ | 2.4 | 2.7 | 2.1 | 3.2 |
| $C_3H_8$ | 1.6 | 1.7 | 1.5 | 3.0 |
| $C_4H_{10}$ | 2.3 | 2.4 | 2.0 | 3.0 |
| CO | 0.1 | 0.5 | 0.2 | 0.5 |
| $H_2S$ | 1.9 | 2.4 | 1.7 | 1.8 |
| $H_2O$ | 8.8 | 12.2 | 6.0 | 7.3 |
| $NH_3$ | 1.1 | 1.1 | 0.9 | 1.1 |
| Oil | 52.1 | 59.1 | 49.8 | 53.2 |
| Asphaltenes | 13.9 | 7.2 | 15.3 | 12.9 |
| Preasphaltenes | 6.9 | 3.0 | 11.3 | 4.2 |
| Unreacted Coal | 6.7 | 5.7 | 7.1 | 6.4 |
| **D. $H_2$ CONSUMPTION, WT. % maf COAL** | 8.1 | 9.1 | 7.3 | 8.4 |

## OPERATING CONDITIONS AND PERFORMANCE FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

| RUN NO. | 8015 | 8016 |
|---|---|---|
| COAL TESTED | DONKIN HR-8 | DONKIN HR-8 |
| CATALYST TESTED | AMOCAT 1C | AMOCAT 1C |
| **A. OPERATING CONDITIONS** | | |
| Unit back pressure, MPa | 17.24 | 17.24 |
| Temperatures, °C: | | |
| First-Stage Reactor | 420 | 420 |
| Second-Stage Reactor | 425 | 425 |
| Hot Separator | 300 | 300 |
| Feedrates: | | |
| Slurry, kg/hr | 0.999 | 1.000 |
| Raw Coal, kg/hr | 0.280 | 0.280 |
| Hydrogen, L/hr | 612. | 612. |
| SHSV, $hr^{-1}$; First-Stage | 1.00 | 1.00 |
| Second-Stage | 0.33 | 0.33 |
| Hydrogen Treatrate, Wt. % Dry Coal | 20.1 | 20.0 |
| Catalyst Age, (kg coal/kg catalyst): | | |
| First-Stage Reactor | 486 | 518 |
| Second-Stage Reactor | 141 | 150 |
| **B. CONVERSIONS OF maf COAL, WT. %** | | |
| Pentane Solubles plus Gases | 69.4 | 69.1 |
| Toluene Solubles plus Gases | 84.9 | 85.2 |
| THF Solubles plus Gases | 93.0 | 93.1 |
| **C. PRODUCT YIELDS ON maf COAL, WT. %** | | |
| $CH_4$ | 2.5 | 2.8 |
| $C_2H_6$ | 2.5 | 2.9 |
| $C_3H_8$ | 2.0 | 2.1 |
| $C_4H_{10}$ | 2.6 | 2.6 |
| CO | 0.2 | 0.2 |
| $H_2S$ | 2.0 | 1.9 |
| $H_2O$ | 7.1 | 7.1 |
| $NH_3$ | 1.0 | 1.0 |
| Oil | 49.5 | 48.5 |
| Asphaltenes | 15.5 | 16.1 |
| Preasphaltenes | 8.1 | 7.9 |
| Unreacted Coal | 7.0 | 6.9 |
| **D. $H_2$ CONSUMPTION, WT. % maf COAL** | 7.9 | 7.8 |

# APPENDIX E

## MATERIAL BALANCES FOR

## CATALYTIC TWO-STAGE LIQUEFACTION EXPERIMENTS

## MATERIAL BALANCES FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

| RUN NO. | 8002 | 8003 | 8004 | 8005 |
|---|---|---|---|---|
| COAL TESTED | LINGAN HR-6 | PRINCE HB-8 | DONKIN HR-8 | LINGAN HR-8 |
| CATALYST TESTED | AMOCAT 1C | AMOCAT 1C | AMOCAT 1C | AMOCAT 1C |
| MOISTURE, WT. % | 1.13 | 3.22 | 1.83 | 1.02 |
| ASH, WT. % DRY BASIS | 4.41 | 5.86 | 5.57 | 2.73 |
| HOURS ON STREAM | 24.0 | 12.0 | 10.0 | 10.0 |
| DURATION OF BALANCE PERIOD, HRS. | 6.0 | 2.1 | 2.0 | 2.0 |
| INPUT (g): | | | | |
| $H_2O$ | 29.3 | 18.6 | 10.7 | 6.0 |
| Ash | 113.2 | 32.7 | 31.9 | 15.9 |
| maf Coal | 2454. | 526. | 540. | 565. |
| Solvent | 6677. | 1485. | 1498. | 1509. |
| $H_2$ | 249.5 | 110.7 | 110.2 | 111.2 |
| Total | 9523. | 2173. | 2191. | 2207. |
| OUTPUT (g): | | | | |
| $H_2O$ | 157.4 | 63.2 | 49.9 | 31.8 |
| Ash | 97.3 | 21.7 | 24.1 | 9.7 |
| Organic THF Insolubles[1] | 232.4 | 40.9 | 38.4 | 56.7 |
| THF Solubles[2] | 8654. | 1890. | 1933. | 1952. |
| $H_2$ | 123.0 | 66.8 | 69.2 | 73.7 |
| Other Gases | 188.6 | 57.9 | 62.7 | 53.7 |
| Total | 9453. | 2141. | 2177. | 2178. |
| TOTAL MATERIAL RECOVERY, WT. % | 99.3 | 98.5 | 99.4 | 98.7 |

1 - Unreacted Coal
2 - Oil, Asphaltenes and Preasphaltenes

## MATERIAL BALANCES FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

| RUN NO. | 8006 | 8007 | 8008 | 8009 |
|---|---|---|---|---|
| COAL TESTED | LINGAN HR-8 | DONKIN HR-8 | PRINCE HB-8 | PRINCE HB-8 |
| CATALYST TESTED | AMOCAT 1C | AMOCAT 1C | AMOCAT 1C | AMOCAT 1C |
| MOISTURE, WT. % | 1.02 | 1.83 | 3.22 | 3.22 |
| ASH, WT. % DRY BASIS | 2.73 | 5.57 | 5.86 | 5.86 |
| HOURS ON STREAM | 10.06 | 10.05 | 10.07 | 10.12 |
| DURATION OF BALANCE PERIOD, HRS. | 2.04 | 2.00 | 2.00 | 1.50 |
| INPUT (g): | | | | |
| $H_2O$ | 6.0 | 10.4 | 18.5 | 13.5 |
| Ash | 16.0 | 31.2 | 32.6 | 23.8 |
| maf Coal | 570. | 529. | 524. | 383. |
| Solvent | 1524. | 1467. | 1480. | 1080. |
| $H_2$ | 113.5 | 110.2 | 110.2 | 82.6 |
| Total | 2230. | 2148. | 2165. | 1583. |
| OUTPUT (g): | | | | |
| $H_2O$ | 30.9 | 48.7 | 63.9 | 50.4 |
| Ash | 8.8 | 20.9 | 19.4 | 15.1 |
| Organic THF Insolubles[1] | 50.7 | 33.1 | 37.7 | 29.4 |
| THF Solubles[2] | 1922. | 1888. | 1873. | 1371. |
| $H_2$ | 70.5 | 64.2 | 65.1 | 48.8 |
| Other Gases | 67.5 | 65.7 | 67.0 | 43.9 |
| Total | 2150. | 2121. | 2126. | 1559. |
| TOTAL MATERIAL RECOVERY, WT. % | 96.5 | 98.7 | 98.2 | 98.5 |

1 - Unreacted Coal
2 - Oil, Asphaltenes and Preasphaltenes

## MATERIAL BALANCES FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

| RUN NO. | 8010 | 8011 | 8012 | 8014 |
|---|---|---|---|---|
| COAL TESTED | DONKIN HR-8 | ILLINOIS NO. 6 | DONKIN HR-8 | DONKIN HR-8 |
| CATALYST TESTED | AMOCAT 1C | AMOCAT 1C | AMOCAT 1C | AMOCAT 1C |
| MOISTURE, WT. % | 1.83 | 3.03 | 1.83 | 1.83 |
| ASH, WT. % DRY BASIS | 5.57 | 11.16 | 5.57 | 5.57 |
| HOURS ON STREAM | 10.08 | 10.05 | 10.07 | 10.02 |
| DURATION OF BALANCE PERIOD, HRS. | 2.00 | 2.00 | 2.02 | 2.00 |
| INPUT (g): | | | | |
| $H_2O$ | 10.5 | 17.6 | 10.9 | 10.6 |
| Ash | 31.5 | 63.0 | 32.4 | 31.6 |
| maf Coal | 534. | 501. | 550. | 535. |
| Solvent | 1480. | 1497. | 1525. | 1485. |
| $H_2$ | 110.2 | 109.1 | 111.3 | 111.2 |
| Total | 2166. | 2188. | 2230. | 2173. |
| OUTPUT (g): | | | | |
| $H_2O$ | 57.7 | 78.7 | 44.1 | 49.8 |
| Ash | 19.6 | 53.8 | 21.9 | 21.7 |
| Organic THF Insolubles[1] | 35.6 | 28.6 | 39.2 | 34.4 |
| THF Solubles[2] | 1894. | 1837. | 1994. | 1908. |
| $H_2$ | 66.9 | 63.7 | 71.4 | 66.2 |
| Other Gases | 61.3 | 64.2 | 57.4 | 85.2 |
| Total | 2135. | 2126. | 2228. | 2165. |
| TOTAL MATERIAL RECOVERY, WT. % | 98.6 | 97.2 | 99.9 | 99.6 |

1 - Unreacted Coal
2 - Oil, Asphaltenes and Preasphaltenes

## MATERIAL BALANCES FOR CATALYTIC TWO-STAGE LIQUEFACTION TESTS

| RUN NO. | 8015 | 8016 |
|---|---|---|
| COAL TESTED | DONKIN HR-8 | DONKIN HR-8 |
| CATALYST TESTED | AMOCAT 1C | AMOCAT 1C |
| MOISTURE, WT. % | 1.83 | 1.83 |
| ASH, WT. % DRY BASIS | 5.57 | 5.57 |
| HOURS ON STREAM | 10.05 | 10.07 |
| *DURATION OF BALANCE PERIOD, HRS.* | 2.00 | 2.00 |
| INPUT (g): | | |
| $H_2O$ | 10.2 | 10.2 |
| Ash | 30.6 | 30.6 |
| maf Coal | 519. | 519. |
| Solvent | 1439. | 1440. |
| $H_2$ | 110.2 | 110.2 |
| Total | 2109. | 2110. |
| OUTPUT (g): | | |
| $H_2O$ | 47.1 | 47.3 |
| Ash | 21.7 | 20.4 |
| Organic THF Insolubles[1] | 36.3 | 35.6 |
| THF Solubles[2] | 1877. | 1882. |
| $H_2$ | 69.3 | 69.9 |
| Other Gases | 65.8 | 69.6 |
| Total | 2117. | 2125. |
| TOTAL MATERIAL RECOVERY, WT. % | 100.4 | 100.7 |

1 - Unreacted Coal
2 - Oil, Asphaltenes and Preasphaltenes

# APPENDIX F

## RESULTS OF HPLC APPLIED TO COAL-DERIVED LIQUIDS

APPENDIX F

## RESULTS OF HPLC APPLIED TO COAL-DERIVED LIQUIDS

In this study, high performance liquid chromatography (HPLC) was used to separate coal liquids into saturates, aromatics and polar compounds. Details pertaining to the HPLC method have been given in an earlier report (6).

Aromatics are separated into "double bond" fractions using this method. A number of model compounds were used to define the specific regions of the chromatograph as shown in Figure 10. The regions were defined by the number of double bonds in a given hydrocarbon molecule ranging from 0 (saturates) to 8 (tetracyclic PAH). Elution profiles for hydrogenated anthracene oil and the pentane soluble fraction of $C_5^+$ liquids from selected two-stage coal liquefaction experiments are shown in Figures 11 - 16. The $C_5^+$ liquids were obtained by combining separator overheads light oil and separator bottoms liquid recovered by vacuum filtration at 75°C.

Results of HPLC analysis presented in Table 17 show that pentane soluble oil from Run Nos. 8003, 8004, 8005 and 8011 contains more saturates and less aromatics than the reaction solvent, HAO 81. Furthermore, the product oils contain a higher level of polar compounds than HAO 81.

Hydrocarbon group analysis of $C_5^+$ liquid product from RCL-7020 and RCL-8000 are shown in Table 18. On day six of the recycle liquefaction test RCL-7020, the product oil contained 26.73% saturates, 61.26% aromatics and 12.01% polar compounds. By day eleven, RCL-8000. saturates in the product oil had declined to 15.00% whereas polar compounds increased to 22.85%. This trend in hydrocarbon group composition is primarily attributed to coke deposition and catalyst fouling which occurred in the second-stage reactor after day six.

Figure 10 - HPLC ELUTION PROFILES FOR MODEL COMPOUNDS

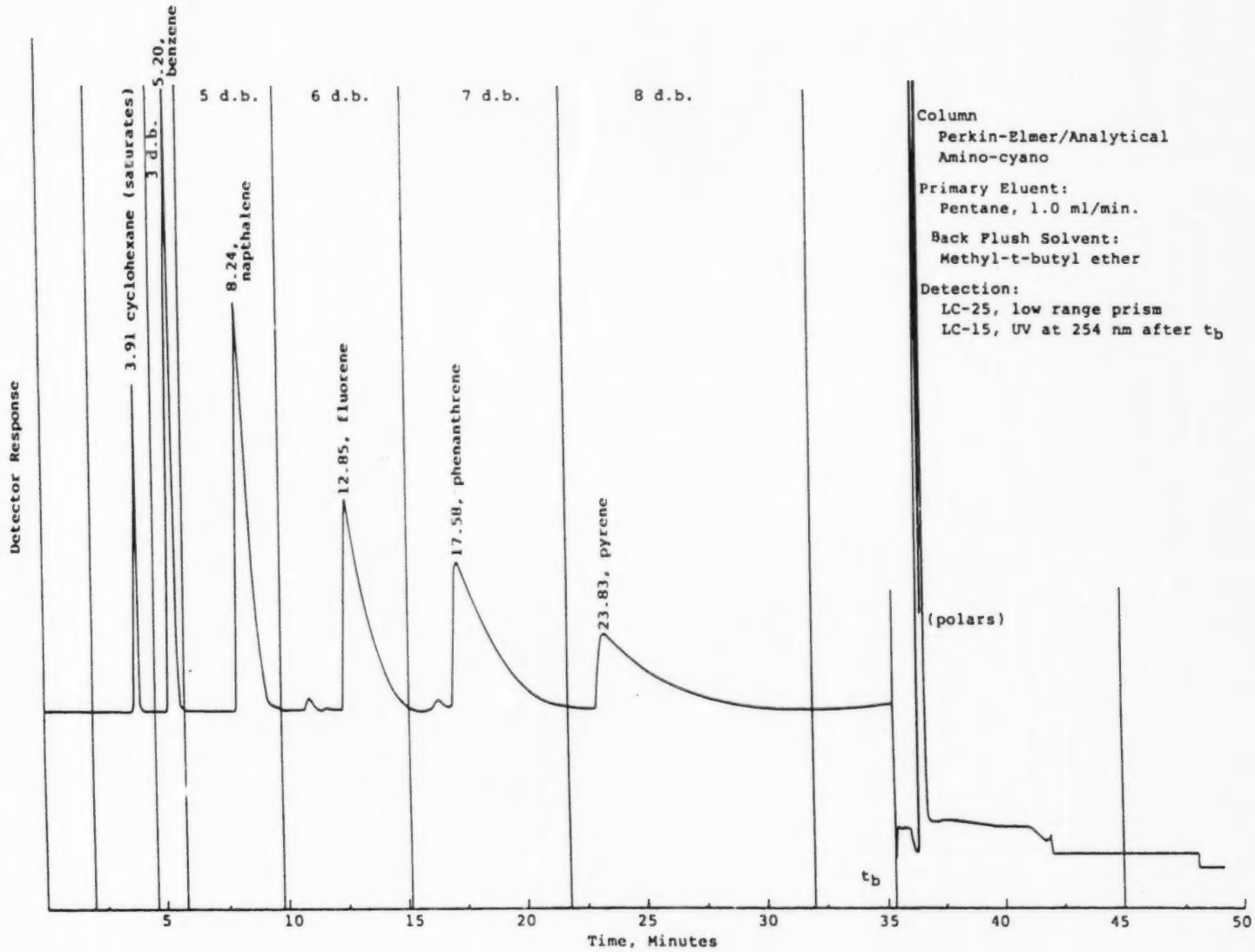

Figure 11 - HPLC SEPARATION OF HYDROGENATED ANTHRACENE OIL WITH 9.02 WT.% OF HYDROGEN

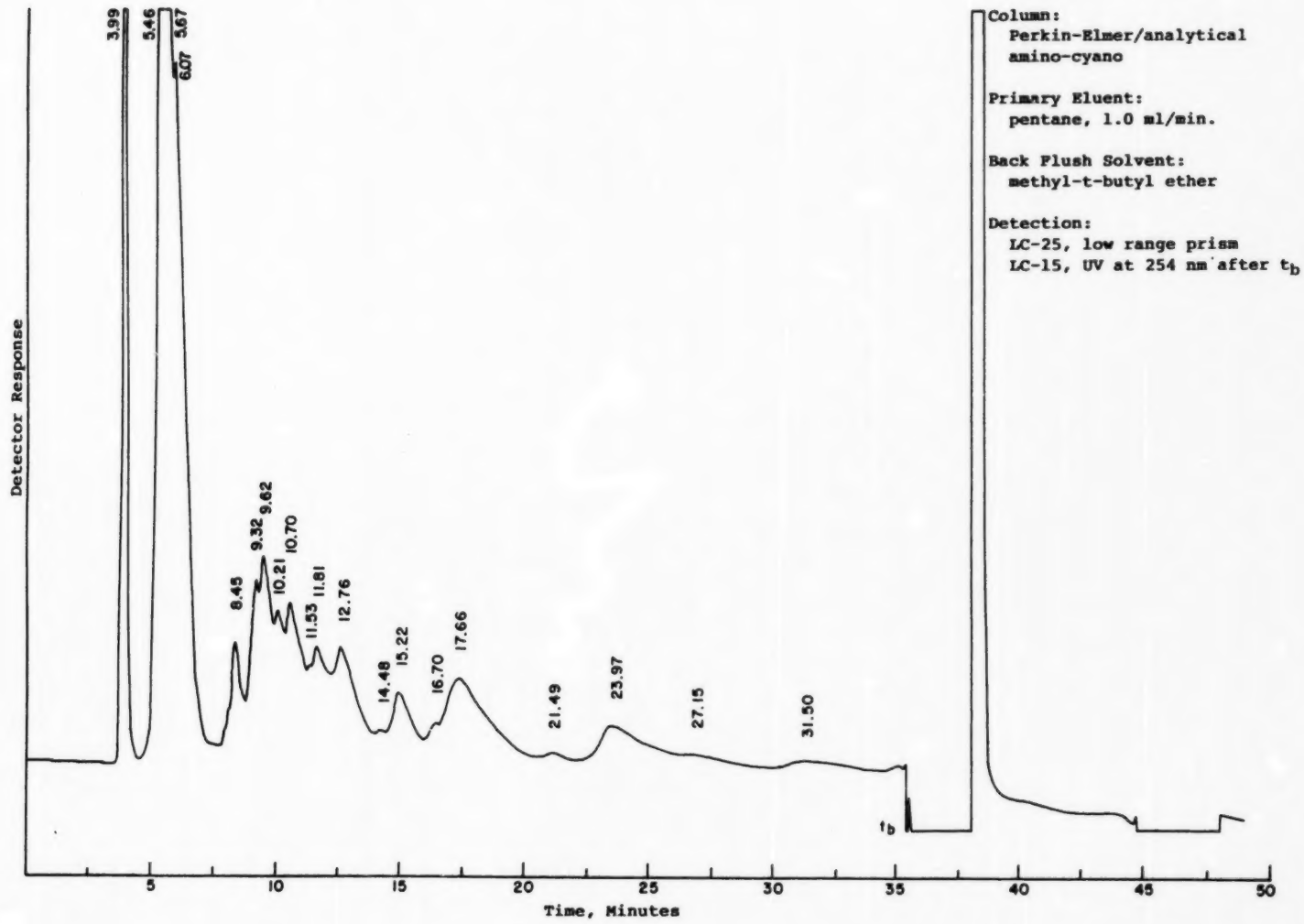

Figure 12 - HPLC SEPARATION OF PENTANE SOLUBLE FRACTION OF $C_5^+$ LIQUID FROM RUN NO. 8003

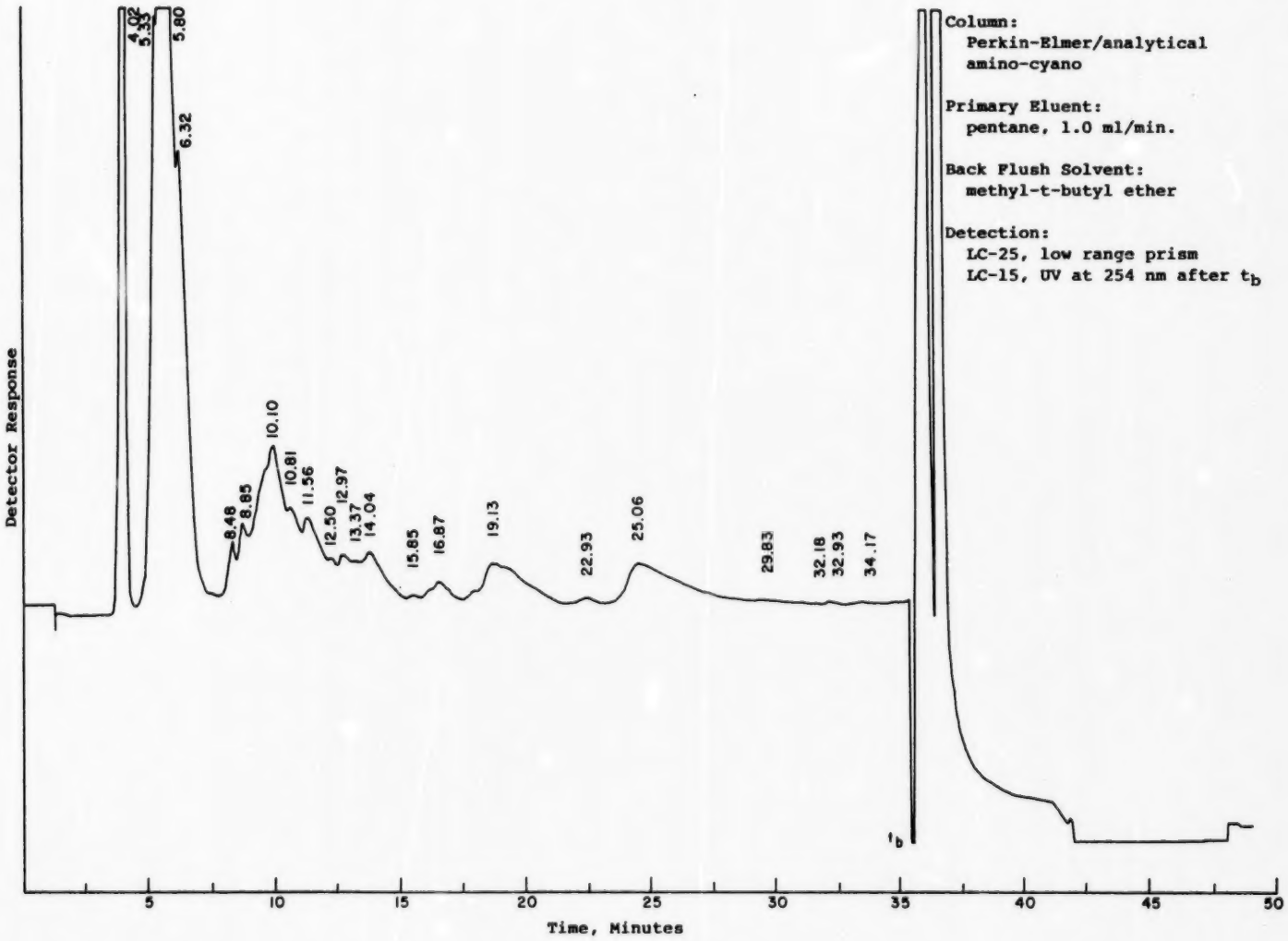

Figure 13 - HPLC SEPARATION OF PENTANE SOLUBLE FRACTION OF $C_5^+$ LIQUID FROM RUN NO. 8004

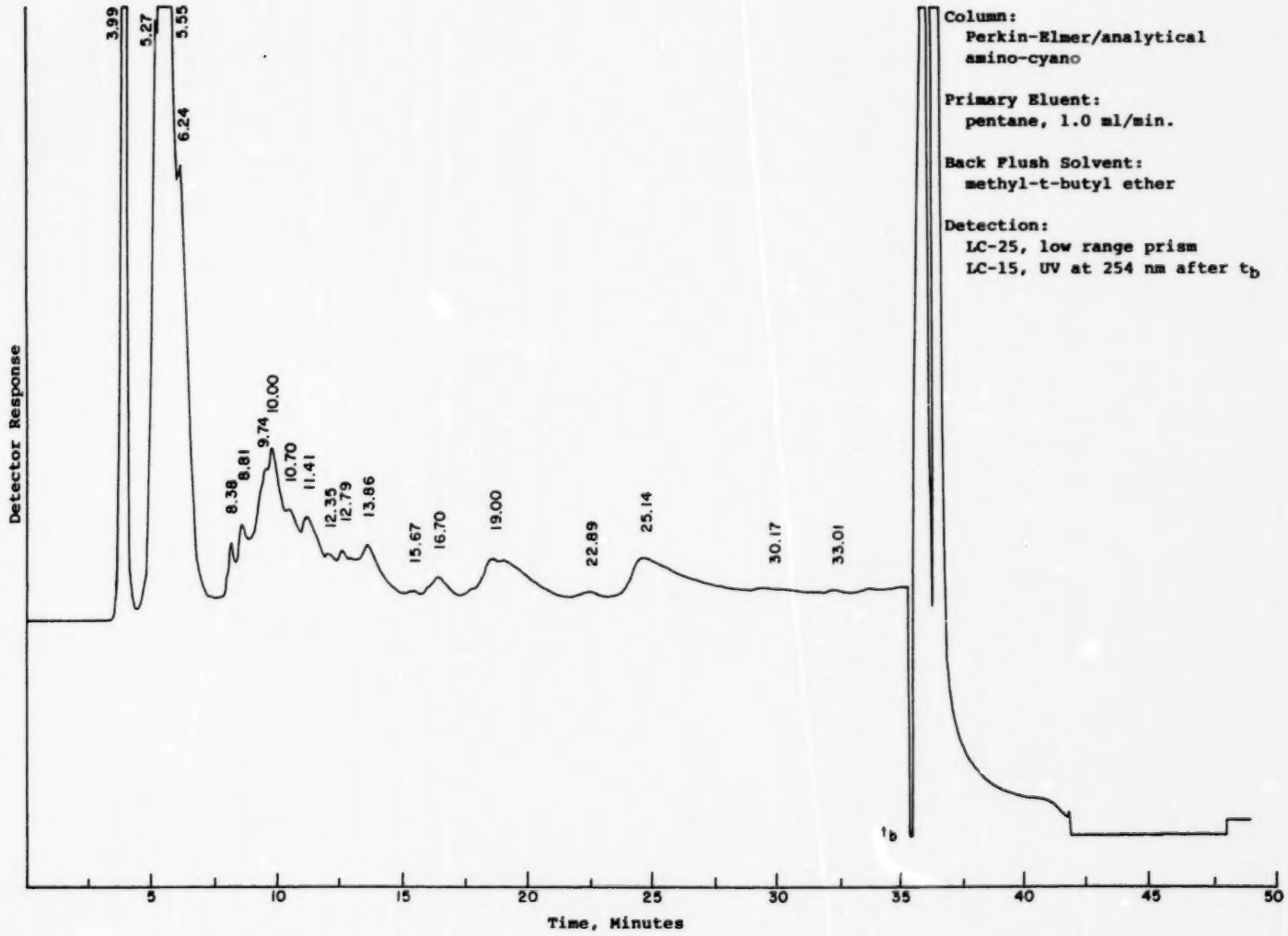

Figure 14 - HPLC SEPARATION OF PENTANE SOLUBLE FRACTION OF $C_5^+$ LIQUID FROM RUN NO. 8005

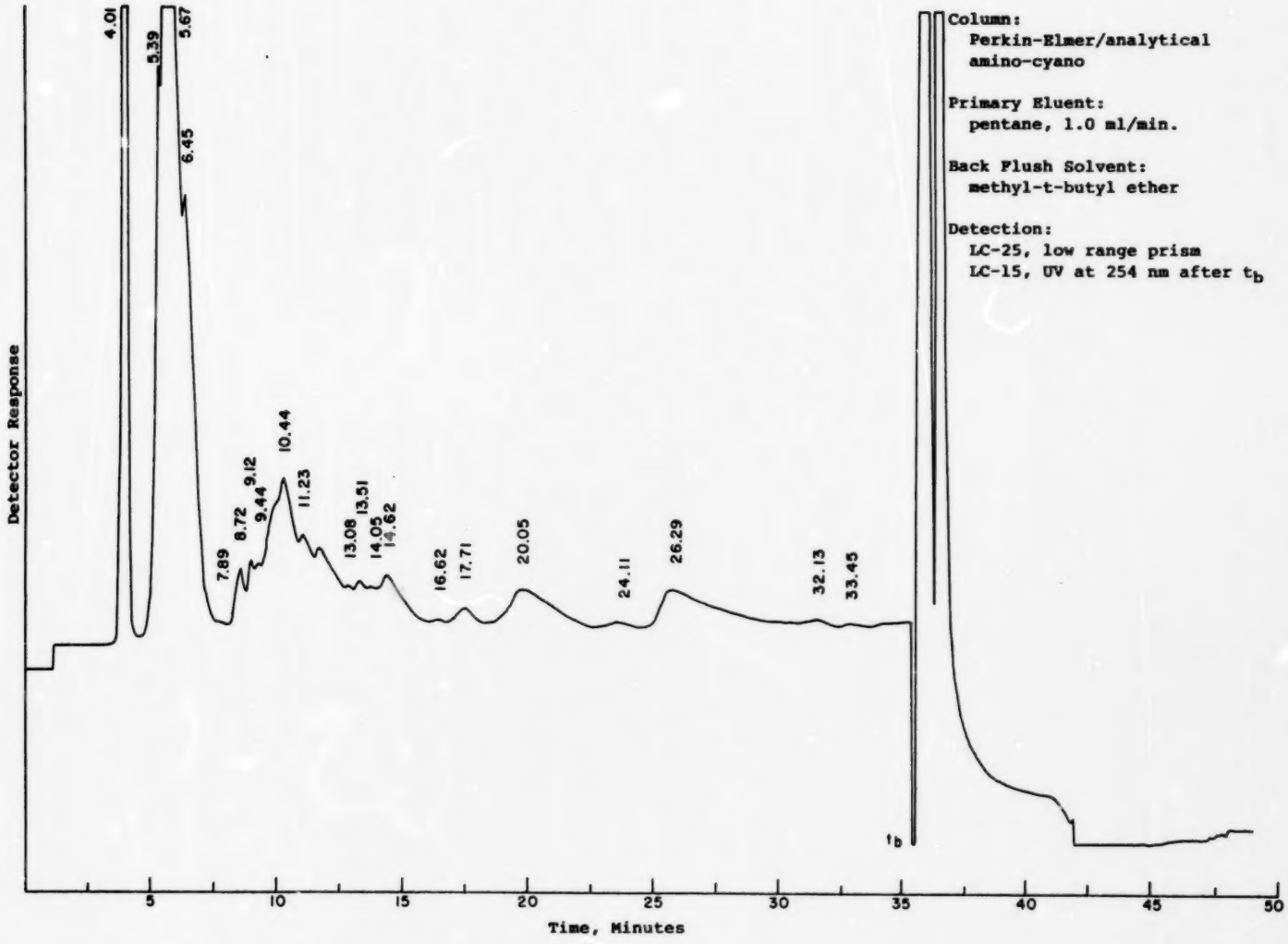

Figure 15 - HPLC SEPARATION OF PENTANE SOLUBLE FRACTION OF $C_5^+$ LIQUID FROM RUN NO. 8011

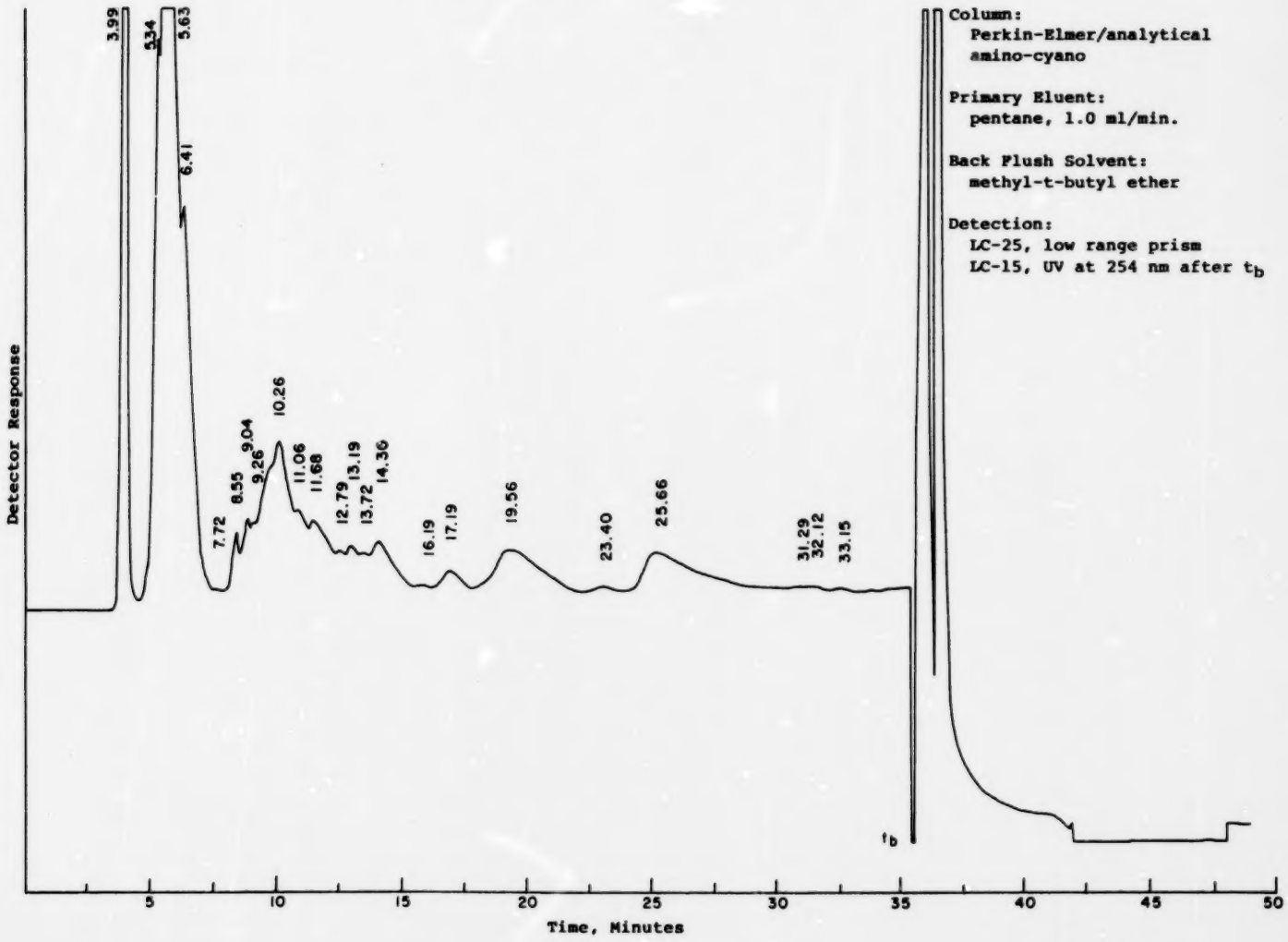

Figure 16 - HPLC SEPARATION OF PENTANE SOLUBLE FRACTION OF $C_5^+$ LIQUID FROM RCL-8000

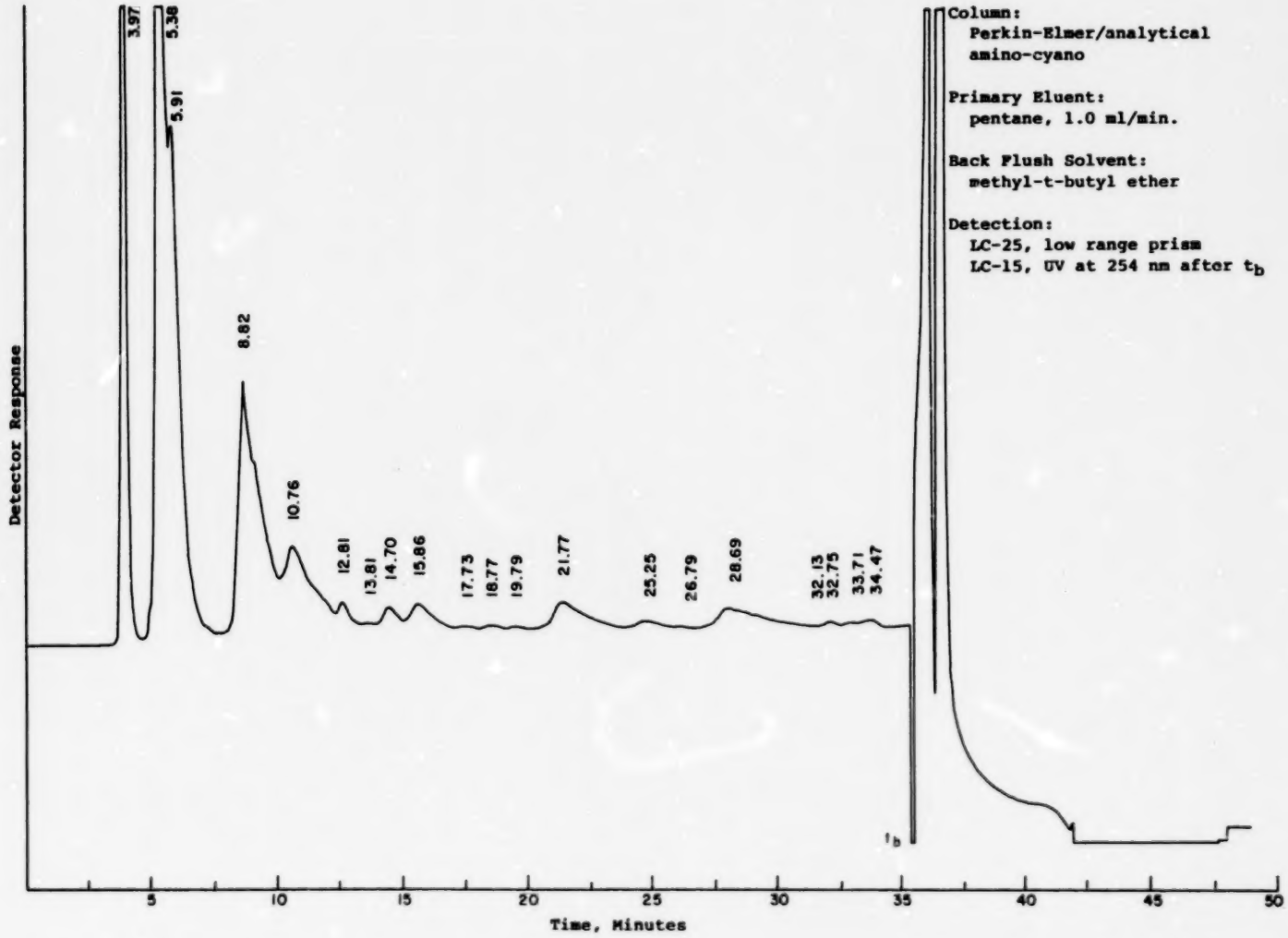

TABLE 17

HPLC SEPARATION OF HYDROGENATED ANTHRACENE OIL AND THE PENTANE SOLUBLE FRACTION OF $C_5^+$ LIQUIDS FROM SELECTED COAL LIQUEFACTION EXPERIMENTS

| SAMPLE DESCRIPTION | HAO 81 | RUN NO. 8003 PRINCE HR-8 | RUN NO. 8004 DONKIN HR-8 | RUN NO. 8005 LINGAN HR-8 | RUN NO. 8011 ILLINOIS NO. 6 |
|---|---|---|---|---|---|
| Specific Gravity, 60/60°F | 1.0294 | 1.0145 | 1.0135 | 1.0178 | 1.0065 |
| Carbon, Wt. % | 90.27 | 89.81 | 90.07 | 90.01 | 90.02 |
| Hydrogen, Wt. % | 9.02 | 9.42 | 9.43 | 9.24 | 9.48 |
| Nitrogen, Wt. % | 0.15 | 0.20 | 0.20 | 0.24 | 0.19 |
| Sulphur, Wt. % | 0.006 | 0.019 | 0.030 | 0.012 | 0.032 |
| Oxygen, Wt. % | 0.26 | 0.31 | 0.31 | 0.17 | 0.24 |
| Pentane Insolubles, Wt. % | NIL | 3.38 | 4.04 | 5.13 | 1.73 |
| HPLC DATA FOR PENTANE SOLUBLE FRACTION: | | | | | |
| Saturates, Wt. % | 13.64 | 19.28 | 18.48 | 18.99 | 19.34 |
| Aromatics, Wt. % | 80.19 | 63.72 | 67.66 | 67.36 | 69.51 |
| 3 double bonds | 12.98 | 20.91 | 25.09 | 21.13 | 26.23 |
| 5 double bonds | 25.65 | 18.96 | 20.79 | 20.97 | 19.02 |
| 6 double bonds | 23.38 | 11.43 | 10.89 | 11.55 | 11.48 |
| 7 double bonds | 9.42 | 4.90 | 4.95 | 5.78 | 5.57 |
| 8 double bonds | 8.76 | 7.52 | 5.94 | 7.93 | 7.21 |
| Polars, Wt. % | 6.17 | 17.00 | 13.86 | 13.65 | 11.15 |

TABLE 18

HPLC SEPARATION OF PENTANE SOLUBLE FRACTION
OF $C_5^+$ LIQUID PRODUCT FROM RCL-7020 AND RCL-8000

| | RCL-7020 | RCL-8000 |
|---|---|---|
| Specific Gravity, 60/60°F | 0.8976 | 0.9826 |
| Carbon, Wt. % | 89.29 | 89.18 |
| Hydrogen, Wt. % | 10.76 | 8.60 |
| Nitrogen, Wt. % | 0.24 | 0.60 |
| Sulphur, Wt. % | 0.071 | 0.100 |
| Oxygen, Wt. % | 0.78 | 1.31 |
| Pentane Insolubles, Wt. % | 3.01 | 10.26 |
| HPLC DATA FOR PENTANE SOLUBLE FRACTION: | | |
| Saturates, Wt. % | 26.73 | 15.00 |
| Aromatics, Wt. % | 61.26 | 62.15 |
| 3 double bonds | 12.38 | 15.72 |
| 5 double bonds | 22.24 | 15.72 |
| 6 double bonds | 13.06 | 10.71 |
| 7 double bonds | 6.35 | 8.57 |
| 8 double bonds | 7.23 | 11.43 |
| Polars, Wt. % | 12.01 | 22.85 |

# APPENDIX G

## OPERATING PROCEDURE FOR

## TWO-STAGE CONTINUOUS COAL LIQUEFACTION UNIT

## PREPARATION OF UNIT PRIOR TO ACTUAL OPERATION

This procedure is normally carried out the day before a run.

1. Open valves 1, 7, 11, 14, 15, 116, 119 and 122.
2. Depressure the system to 345 kPa by opening valves 17 and 18. Close valves 17 and 18.
3. Open valve 2 and valve 8 and pressurize the system as well as the 7.6 liter emergency blowdown vessel to 4.14 MPa. Close valve 8 and continue pressurizing the system until the pressure is at the desired level either 10.34 MPa or 13.79 MPa. Close valve 2.
4. Leave for a few hours and monitor pressure.
5. Set the forward pressure regulator and the back pressure regulator to the desired settings.
6. Ensure that alarm switch No. 14 is deactivated.

## ACTUAL OPERATION

### Start-Up

1. Turn on the cooling water to the magnedrive stirrers.
2. Set the controllers at the desired settings for both the first-stage and second-stage reactors.

| | Stage 1 | Stage 2 |
|---|---|---|
| reactor controller, °C | 343 | 288 |
| furnace controller, °C | 343 | 316 |
| zone controllers, % | 60 | 100 |

3. Turn on the temperature controller for high pressure separator No. 1, V-5, at 300°C.
4. Turn on the temperature controller for high pressure separator No. 2, V-6, at 300°C.
5. Gradually increase the furnace controllers for stage 1 and stage 2 to the desired levels. NEVER allow the heating rate of the reactors to exceed 120°C per hour.
6. After two hours of heat-up time follow steps 7, 8 and 9.

7. Turn on heating tapes on slurry transfer lines:

   No. 1 - 33%, at start of run gradually increase to 100%

   No. 2 - 33%, at start of run gradually increase to 66%

   No. 3 - 33%

   No. 4 - 33%

8. Turn on condenser cooling water.

9. Activate K-Ray electronics for liquid level measurement system.

10. After one hour open the window on the radioactive source.

11. Turn on the hydrogen flow control system fifteen minutes prior to run.

Procedure

a) Turn on the power.
b) Open valve 3.
c) Set the air pressure at 138 kPa.
d) Adjust the setpoint knob until the ball is in the center, then switch from manual to automatic.
e) Set the pen to the desired value.
f) Open valve 123 and close valve 122.
g) Allow the hydrogen flow control system to stabilize.
h) Monitor exit gas flow rate for 15 minutes.

12. Purge the feed tank with nitrogen 15 minutes prior to slurry addition. Maintain a small nitrogen flow throughout the run.

13. Charge V-1 with the slurry. Open valve 101 and turn on the circulating pump.

14. Turn off the circulating pump for 15 seconds and then turn the circulating pump back on. This fills void spaces or gas pockets in the pump with slurry and ensures proper weight readings on the toledo scale. Turn on the alarm switch No. 14 in the side panel.

15. When the temperatures of the reactors are at the desired levels open valve 102 and purge the high pressure metering pump with slurry until the liquid is gas free.

Run

16. Open the plant valve (valve 5 or 6) and activate the high pressure metering pump.

17. Turn on the V-3, V-4 and V-5 magnedrive stirrers.

18. Turn on the cooling fan for the magnedrive stirrer on V-4.

19. Monitor the pressures, temperatures, slurry feedrate and vent gas flow rates throughout the run.

20. A rapid increase in the reactor 1 temperature will occur 10 to 20 minutes after the high pressure metering pump has been activated. When the reactor temperature reaches 4°C below the target temperature turn the furnace temperature controller down until the red light goes off. Increase the furnace temperature controller back to the original setpoint when reactor 1 temperature levels off.

21. Open valves 111, 107, 104 and 105 and purge V-9 with nitrogen, 10 minutes prior to step 22.

22. After 2.3 kg of slurry has been pumped, open valve 22.

23. Set timer K-5 for the automatic letdown (ALD) system to maintain a constant level in V-5.

24. Open valve 19.

25. Set the selector switch on "Automatic During High Alert" and activate the automatic letdown system.

26. After the first ALD cycle (mark this on the data sheet) close valve 105.

27. Sample vent gas streams 15 minutes prior to the steady state sampling period.

28. Drain liquid from the separator overheads knock-out (V-7) periodically and 5 minutes prior to the steady state sampling period by opening and closing valves 17 and 18 sequentially.

29. To initiate the steady state sampling period, record the toledo scale weight after valve "A" has closed on the final ALD cycle of the slop period.

30. Record the wet test meter reading and the circulating and metering pump pan weights.

31. Open valves 108 and 23. Close valves 22 and 107. Open valve 106 and start a nitrogen purge.

32. On the next ALD cycle, the first in the steady state sampling period, mark the stroke made on the hydrogen flow recorder.

33. Close valve 106.

34. Open valve 120 and drain V-15. Close valve 120.

35. Sample vent gas streams 15 minutes prior to the end of the steady state sampling period.

36. Five minutes prior to end of run, drain liquid from separator overheads knock-out, V-7, by opening and closing valves 17 and 18 sequentially.

37. At the end of the period, record the toledo scale weight after valve "A" has closed on the final ALD cycle of the sampling period.

38. Open valves 107 and 22. Close valves 23 and 108. Open valve 105 and start a nitrogen purge. After the next ALD cycle close valve 105.

39. Record the wet test meter reading and the circulating and metering pump pan weights. Empty contents of V-15.

40. Turn the reactor controller setpoints on stage 1 and stage 2 reactors down to 260°C. Turn the furnace controller setpoints on stage 1 and stage 2 reactors down to 204°C. Reduce voltage to heating tapes to 33% on slurry transfer lines. Turn off high pressure separator heaters.

41. Record the toledo scale weight and deactivate the circulating pump for 15 seconds. Reactivate the circulating pump and record the toledo scale weight.

42. Turn down the hydrogen flow control system to ca. 100 S/D.

43. Deactivate the high pressure metering pump.

44. Close the plant valve (valve 5 or 6) and valves 11 and 14.

45. Deactivate the circulating pump and drain slurry from the feed tank.

46. Close valves 101 and 102 and add flush oil to V-1, open valve 101 and turn on the circulating pump.

Flush Cycle and Shutdown

47. Open the priming valve above the metering pump and allow 1 kg of flush oil to exit. Close the priming valve.

48. Open the plant valve and activate the high pressure metering pump.

49. Reactivate the reactor controller setpoints in both reactors to 371°C. Reactivate the furnace controller setpoints in both reactors to ca. 540°C. Turn on the heater on separator No. 1 to 300°C. Increase voltage on line heaters to 66% for the preheater and 33% for the interstage and slurry transfer lines. Increase the hydrogen flow control setpoint to 300 S/D.

50. Note: Keep temperatures of stage 1 and stage 2 reactors between 343°C and 371°C during flush cycle.

51. Open valve 111 and 120 and drain V-15. Close valve 111.

52. When approximately 3.0 kg of oil has been pumped into V-5, open valves 111, 108, 23 and 19. Activate the ALD system allowing it to operate until at least 14 kg of flush oil has been pumped. Close valve 19, allow one more cycle. Deactivate the ALD system and close valves 23, 108 and 111.

53. Pull the plug out for electronics on the ALD system and close the window on the radioactive source. Shut off hydrogen control system.

Procedure

a) Turn the regulator knob until the ball is in the centre.
b) Switch from automatic to manual.
c) Shut off the air flow.
d) Close valve 3.
e) Turn off the power.

54. Deactivate the metering pump.

55. Shut off all heating tapes, heaters and the reactor furnaces.

56. Turn off the condenser cooling water. Leave the magnedrive water on overnight.

57. Close the plant valve and valves 11 and 14.

58. Withdraw oil from V-5.

59. Open valves 14 and 15 to equalize the pressure in V-5, V-6 and V-7.

60. Open valve 11 to equalize the pressure throughout the system.

61. Close the interstage valve and valve 11.

62. Transfer contents of V-4 into the 7.6 liter high pressure vessel by opening valve 9. Let stand for 5 minutes then close valve 9. Ensure that pressure in V-4 stays above 6.89 MPa.

63. Transfer contents of V-3 into the 7.6 liter high pressure vessel by opening valve 8. Let drain for 5 minutes.

64. Open valve 11 and equalize pressure in V-4, V-5 and V-6.

65. Open valve 2 and add high pressure hydrogen to V-3 until its pressure is 689 kPa in excess of V-4. Close valve 2.

66. Open the interstage valve (valve 7) to equalize system pressure.

67. Open valve 2 and add hydrogen to the system until the pressure is 11.38 MPa.

68. Drain flush oil from V-1.

69. Turn off electronics for the toledo scale and the digital pressure meter.

70. Shut off alarm switch No. 14 in the side panel.

## POST RUN OPERATIONS

This procedure is normally carried out the day after the run.

1. Open valve 2 and pressurize the system to 10.34 MPa.

2. Close the interstage valve (valve No. 7) and valves 11 and 14.

3. Depressure the 7.6 liter pressure vessel to 4.83 MPa.

4. Drain contents of V-5 by opening valves 110, 26 and 24. Close valves 24, 26 and 110.

5. Open valve 14 and equalize the pressure in V-5, V-6 and V-7.

6. Empty contents of V-4 by opening valve 9. Close valve 9.

7. Depressure the 7.6 liter pressure vessel to 4.83 MPa.

8. Empty contents of V-3 by opening valve 8. Close valve 8.

9. Open valve 11.

10. Follow steps 65 and 66. Then open valve 2 and add hydrogen to the system until the pressure is ca. 12.93 MPa.

11. Depressure the 7.6 liter pressure vessel to 1.38 MPa. Empty contents of the 7.6 liter vessel into the 10 liter low pressure vessel.

12. Follow steps 1 to 4 on page G-2.

SCHEMATIC DIAGRAM OF TWO-STAGE COAL LIQUEFACTION BENCH UNIT